

Spin-Charge Bound States and Emerging Fermions in a Quantum Spin Liquid

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The complex interplay between charge and spin dynamics lies at the heart of strongly correlated quantum materials, and it is a fundamental topic in basic research with far-reaching technological perspectives. We explore in this paper the dynamics of holes in a single-band, extended t - J model where the background spins form a $\mathbb{Z}_2$ quantum spin liquid. Using a field theory approach based on a parton construction, we show that while the electrons for most momenta fractionalize into uncorrelated charge-carrying holons and spin-carrying spinons as generally expected for a quantum spin liquid, the spinon-holon scattering cross section diverges for certain momenta, signaling strong correlations. By deriving an effective low-energy Hamiltonian describing this dynamics, we demonstrate that these divergences are due to the formation of long-lived spinon-holon bound states. Since the wave function of these bound states is localized over a few lattice sites, they correspond to well-defined fermions with the same charge and spin as the underlying electrons. We then show that quantum gas microscopy with atoms in optical lattices provides an excellent platform for verifying and probing the internal spatial structure of these emerging fermions. The fermions will furthermore show up as clear quasiparticle peaks in angle-resolved photoemission spectroscopy with an intensity determined by their internal structure. For a nonzero hole concentration, the fermions form hole pockets with qualitatively the same location, shape, and intensity variation in the Brillouin zone as the so-called Fermi arcs observed in the pseudogap phase. Such agreement is remarkable since the Fermi arcs arise from the delicate interplay between the symmetry of the quantum spin liquid and the internal structure of the emerging fermions in a minimal single-band model with no extra degrees of freedom added. Our results, therefore, provide a microscopic mechanism for the conjectured fractionalized Fermi liquid and open up new pathways for exploring the pseudogap phase and high-temperature superconductivity as arising from a quantum spin liquid.

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I. INTRODUCTION

The reduced dimensionality of two-dimensional (2D) materials renders their properties highly sensitive to quantum mechanical effects. This gives rise to exotic phenomena such as long-range quantum entanglement,

quasiparticles with nontrivial statistics, and spin-charge separation [1]. A prominent example is quantum spin liquids (QSLs) consisting of a macroscopic superposition of spin singlet configurations covering the lattice, an idea which, since it was introduced more than 50 years ago, continues to attract significant attention, with many fundamental and open questions [2–5]. A QSL was recently observed with the use of Rydberg atoms in an optical tweezer array [6], and there is promising progress toward realizing a QSL with atoms in optical lattices [7–11]. Diverse solid-state materials with kagome and triangular lattices show signs of QSL physics [12–14]. QSLs are also predicted to exist in the rapidly growing class of 2D materials consisting of twisted atomically thin layers

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[15–18]. A prevailing paradigm regarding QSLs is that their low-energy excitations have lost any resemblance to the underlying electrons, which instead have “fractionalized” into largely uncorrelated spin and charge degrees of freedom [19,20]. It has, however, been suggested that a new kind of fractionalized Fermi liquid (FL*) can exist in such QSLs when doped away from half-filling. This may, moreover, be connected to the puzzling pseudogap phase observed in the cuprates [21–31], following an early idea that high-temperature superconductivity arises from a QSL [32–36].

Inspired by this, we explore in this paper the properties of holes in a $\mathbb{Z}_2$ quantum spin liquid background described by a single-band, extended t - J model on a square lattice close to half-filling. We develop a field-theoretic framework based on a parton construction, from which we derive an effective low-energy Hamiltonian describing the hole dynamics in terms of charge-carrying holons interacting with spin-carrying spinons. While the spinons and holons are essentially uncorrelated for most center-of-mass (COM) momenta in agreement with the prevailing paradigm for QSLs, we show that they can form long-lived bound states in certain regions in the Brillouin zone (BZ). These bound states have a relative wave function localized over a few lattice sites and, therefore, describe emerging fermions with a well-defined energy, charge, and spin. We then demonstrate that the internal spatial structure of these fermions can be probed with the use of well-established quantum gas microscopy techniques in optical lattices, and that they show up as sharp peaks in angle-resolved photoemission spectroscopy (ARPES) experiments. Remarkably, for a nonzero concentration of holes, they form pockets in the BZ giving rise to ARPES spectra with qualitatively the same position and shape as the intriguing Fermi arcs observed in the pseudogap phase. This shape arises from the subtle interplay between the symmetries of the spin liquid and the internal structure of the fermions emerging from the t - J model with no fitting parameters or extra degrees of freedom added. As such, our theory provides a microscopic basis for the conjectured existence of a FL* state in the pseudogap phase of the cuprates. Our main results are shown in Fig. 1.

This paper is organized as follows. Section II introduces the extended t - J model together with the parton description of holes in the spin liquid. In Sec. III, we discuss our field-theoretic approach to calculate the hole Green’s function, and in Sec. IV we derive an effective Schrödinger equation giving a nonperturbative description of the holon-spinon correlations. The emergence and properties of spinon-holon bound states are discussed in Sec. V, and we explore in Sec. VI how these bound states can be probed with quantum gas microscopy in optical lattices as well as with ARPES in condensed-matter systems. We end with conclusions and an outlook in Sec. VII.

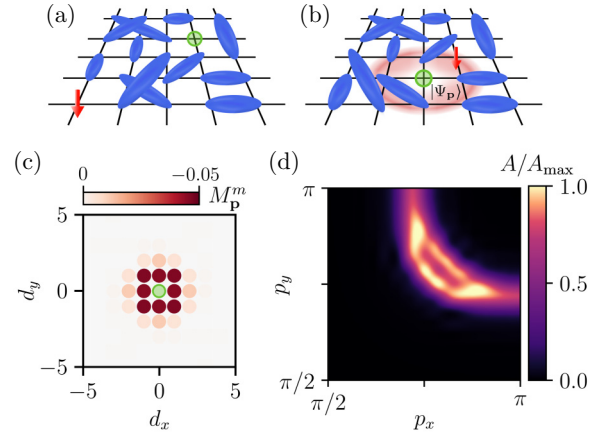


FIG. 1. (a) A physical spin $\uparrow$ hole fractionalizes into an unbound spinon (arrow) and holon (green circle) pair in a spin liquid formed by spin singlets (blue ovals). (b) For some momenta, however, the holon and spinon bind to form a new fermionic quasiparticle with well-defined spin and charge, and a spatial structure determined by the relative spinon-holon wave function $|\Psi_{\mathbf{p}}\rangle$ extending over a few lattice sites as indicated by the red coloring. (c) Mimicking the spatial structure of the bound state, the hole-spin correlation function $M_{\mathbf{p}}^n(\mathbf{d})$ effectively describes the likelihood of finding the unpaired spin at a given lattice with the holon (green circle) at the center. (d) The spectral response in the Brillouin zone of these fermions at their Fermi energy as measured by angle-resolved photoemission spectroscopy for a nonzero hole doping $x = 0.01$, where they form a FL*. This clearly shows a hole pocket around $\mathbf{p} = (3\pi/4, 3\pi/4)$ with a dimly lit back side due to the interplay between the internal symmetry of the fermions and the quantum spin liquid singlets. There are identical hole pockets in the other corners of the Brillouin zone. The shape and spectral weight of these pockets are similar to those of the Fermi arcs observed in the pseudogap phase of the cuprates.

II. MODEL

We consider spin $\sigma = \uparrow, \downarrow$ fermions on a two-dimensional square lattice with nearest-neighbor t_1 and next-nearest-neighbor t_2 hopping and on-site repulsion U . This is described by a single-band Hubbard model, which in the limit of strong repulsion $t_i/U \ll 1$ close to half-filling (one electron per site) reduces to the extended t - J model $\hat{H} = \hat{H}_t + \hat{H}_J$. Here,

$$\hat{H}_t = -t_1 \sum_{\langle \mathbf{i}, \mathbf{j} \rangle, \sigma} (\tilde{c}_{\mathbf{i}, \sigma}^\dagger \tilde{c}_{\mathbf{j}, \sigma} + \text{h.c.}) - t_2 \sum_{\langle\langle \mathbf{i}, \mathbf{j} \rangle\rangle, \sigma} (\tilde{c}_{\mathbf{i}, \sigma}^\dagger \tilde{c}_{\mathbf{j}, \sigma} + \text{h.c.}) \quad (1)$$

is the kinetic energy, with $\langle \mathbf{i}, \mathbf{j} \rangle$ nearest-neighbor and $\langle\langle \mathbf{i}, \mathbf{j} \rangle\rangle$ next-nearest-neighbor lattice sites, with $\tilde{c}_{\mathbf{i}, \sigma}^\dagger = \hat{c}_{\mathbf{i}, \sigma}^\dagger (1 - \hat{n}_{\mathbf{i}})$, and where $\hat{c}_{\mathbf{i}, \sigma}$ removes a fermion with spin σ at site $\mathbf{i}$ so that $\hat{n}_{\mathbf{i}} = \sum_{\sigma} \hat{c}_{\mathbf{i}, \sigma}^\dagger \hat{c}_{\mathbf{i}, \sigma}$ is the density operator. The

antiferromagnetic Heisenberg term is

$$\hat{H}_J = J_1 \sum_{\langle i,j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j, \quad (2)$$

where $\hat{\mathbf{S}}_i = \frac{1}{2} \sum_{\sigma,\sigma'} \hat{c}_{i,\sigma}^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} \hat{c}_{i,\sigma'}$, where $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ are spin operators and $J_i = 4t_i^2/U$. We have in Eq. (2) left out a density-density interaction $-\sum_{\langle i,j \rangle} \hat{n}_i \hat{n}_j/4$, since it becomes a constant for a single hole injected into a half-filled background.

At half-filling the model reduces to the J_1 - J_2 Heisenberg model $\hat{H}_J$, whose ground state is predicted to be a $\mathbb{Z}_2$ QSL in a finite region of the phase diagram for spin couplings around $J_2/J_1 = 0.5$ [37–42]. Our goal is to study the hole dynamics in this QSL phase, and we use a parton construction where the electron operator is decomposed as $\hat{c}_{i,\sigma} = \hat{b}_i^\dagger \hat{f}_{i,\sigma}$. Here, $\hat{f}_{i,\sigma}$ removes a fermionic spinon, which carries spin but no charge, and $\hat{b}_i$ removes a bosonic holon carrying charge but no spin [20]. With the use of this construction, the Heisenberg term $\hat{H}_J$ becomes quartic in the spinon operators and different spin liquids can then be described with the use of different mean-field theories as a starting point [43]. By comparison with results from variational Monte Carlo calculations [37,41], it was found that

$$\hat{H}_J^{\text{mf}} = \sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}} \hat{f}_{\mathbf{k},\sigma}^\dagger \hat{f}_{\mathbf{k},\sigma} + \sum_{\mathbf{k}} \left(\Delta_{\mathbf{k}} \hat{f}_{\mathbf{k},\uparrow}^\dagger \hat{f}_{-\mathbf{k},\downarrow}^\dagger + \text{h.c.} \right) \quad (3)$$

provides a mean-field description of the $\mathbb{Z}_2$ QSL as well as its low-energy excitations [44]. Here,

$$\begin{aligned} \epsilon_{\mathbf{k}} &= \epsilon(\cos k_x + \cos k_y), \\ \Delta_{\mathbf{k}} &= \Delta_1(\cos k_x - \cos k_y) + \Delta_2 \sin(2k_x) \sin(2k_y) \end{aligned} \quad (4)$$

are the dispersion and d_{xy} -wave pairing fields, describing the singlet formation in the QSL, $\mathbf{k}$ is a crystal momentum in the BZ, and the lattice constant is taken to be unity. We have $\Delta_1 = 1.8\epsilon$ and $\Delta_2 = 1.1\epsilon$ for $J_2/J_1 = 0.5$ [41], and from Ref. [44] we estimate $\epsilon \approx J_1/3$. In the following, we fix $J_1/J_2 = 0.5$, such that the only free parameter is the interaction strength t_1/J_1 controlling the competition between the charge motion and spin ordering. We set $t_2/t_1 = -\sqrt{J_2/J_1}$ to describe the case of hole doping; $t_2/t_1 = +\sqrt{J_2/J_1}$ would instead describe electron doping [45].

Diagonalization of Eq. (3) yields

$$\hat{H}_J^{\text{mf}} = \sum_{\mathbf{k},\sigma} \omega_{\mathbf{k}}^s \hat{\gamma}_{\mathbf{k}\sigma}^\dagger \hat{\gamma}_{\mathbf{k}\sigma}, \quad (5)$$

with $\hat{\gamma}_{\mathbf{k}\sigma}$ removing a spinon with momentum $\mathbf{k}$, spin σ , and energy $\omega_{\mathbf{k}}^s = \sqrt{\epsilon_{\mathbf{k}}^2 + \Delta_{\mathbf{k}}^2}$. We have $\hat{f}_{\mathbf{k}\uparrow} = u_{\mathbf{k}} \hat{\gamma}_{\mathbf{k}\uparrow} - v_{\mathbf{k}} \hat{\gamma}_{-\mathbf{k}\downarrow}^\dagger$

and $\hat{f}_{\mathbf{k}\downarrow} = u_{\mathbf{k}} \hat{\gamma}_{\mathbf{k}\downarrow} + v_{\mathbf{k}} \hat{\gamma}_{-\mathbf{k}\uparrow}^\dagger$ with the coherence factors $u_{\mathbf{k}} = \sqrt{(1 + \epsilon_{\mathbf{k}}/\omega_{\mathbf{k}}^s)/2}$ and $v_{\mathbf{k}} = \text{sgn}(\Delta_{\mathbf{k}}) \sqrt{(1 - \epsilon_{\mathbf{k}}/\omega_{\mathbf{k}}^s)/2}$.

When holes are introduced, their motion can destroy the singlets of the QSL. This is described by the hopping term, which reads

$$\begin{aligned} \hat{H}_t &= \sum_{\mathbf{k}} \omega_{\mathbf{k}}^h \hat{b}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}} + \sum_{\mathbf{p},\mathbf{k},\mathbf{q},\sigma} h_{\mathbf{p},\mathbf{k},\mathbf{q}} \hat{b}_{\mathbf{p}-\mathbf{q}}^\dagger \hat{b}_{\mathbf{p}} \hat{\gamma}_{\mathbf{k}+\mathbf{q},\sigma}^\dagger \hat{\gamma}_{\mathbf{k},\sigma} \\ &+ \sum_{\mathbf{p},\mathbf{k},\mathbf{q}} g_{\mathbf{p},\mathbf{k},\mathbf{q}} \left(\hat{b}_{\mathbf{p}-\mathbf{q}}^\dagger \hat{b}_{\mathbf{p}} \hat{\gamma}_{\mathbf{k}+\mathbf{q},\downarrow}^\dagger \hat{\gamma}_{\mathbf{k},\uparrow}^\dagger + \text{h.c.} \right) \end{aligned} \quad (6)$$

in terms of partons [46]. Here

$$\begin{aligned} g_{\mathbf{p},\mathbf{k},\mathbf{q}} &= \Lambda_{\mathbf{p}-\mathbf{k}} u_{\mathbf{k}-\mathbf{q}} v_{\mathbf{k}-\mathbf{q}} + \Lambda_{\mathbf{p}+\mathbf{k}-\mathbf{q}} v_{\mathbf{k}} u_{\mathbf{k}-\mathbf{q}}, \\ h_{\mathbf{p},\mathbf{k},\mathbf{q}} &= \Lambda_{\mathbf{p}-\mathbf{k}-\mathbf{q}} u_{\mathbf{k}+\mathbf{q}} u_{\mathbf{k}} - \Lambda_{\mathbf{p}+\mathbf{k}} v_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}} \end{aligned} \quad (7)$$

are the vertex functions for the holon scattering on a spinon and emitting/absorbing two spinons, and

$$\omega_{\mathbf{p}}^h = 2 \sum_{\mathbf{k}} \Lambda_{\mathbf{p}+\mathbf{k}} v_{\mathbf{k}}^2 \quad (8)$$

is the bare holon dispersion with

$$\begin{aligned} \Lambda_{\mathbf{k}} &= -\frac{2}{N} \left(t_1 [\cos k_x + \cos k_y] \right. \\ &\quad \left. + t_2 [\cos(k_x + k_y) + \cos(k_x - k_y)] \right). \end{aligned} \quad (9)$$

We focus in this paper on the hole spectrum, which can be measured with ARPES as discussed in Sec. VI B. The spectrum related to inserting an extra electron is separated by a large energy gap U since it involves at least one lattice site with double occupancy.

III. FIELD THEORY

We now describe our field-theoretic approach for analyzing the dynamics of holes in the QSL. Removal of a spin $\uparrow$ electron from the QSL at half-filling is described by the retarded hole Green's function

$$\begin{aligned} G(\mathbf{p}, \tau) &= -i\theta(\tau) \langle \{ \hat{c}_{\mathbf{p},\uparrow}^\dagger(\tau), \hat{c}_{\mathbf{p},\uparrow}(0) \} \rangle \\ &= -\frac{i\theta(\tau)}{N} \sum_{\mathbf{q}_1, \mathbf{q}_2} v_{\mathbf{q}_1} v_{\mathbf{q}_2} \left\langle \hat{\gamma}_{\mathbf{q}_2, \downarrow}^\dagger(\tau) \hat{b}_{\mathbf{p}-\mathbf{q}_2}(\tau) \hat{b}_{\mathbf{p}-\mathbf{q}_1}^\dagger \hat{\gamma}_{\mathbf{q}_1, \downarrow}^\dagger \right\rangle, \end{aligned} \quad (10)$$

where $\hat{A}(\tau) = \exp(i\hat{H}\tau) \hat{A} \exp(-i\hat{H}\tau)$ is an operator at time τ in the Heisenberg picture. As is apparent from Eq. (10), the one-body Green's function for a physical hole is a *two-body* spinon-holon Green's function in the parton picture, since the removal of a spin $\uparrow$ electron creates a holon and a spin $\downarrow$ spinon. Calculation of this

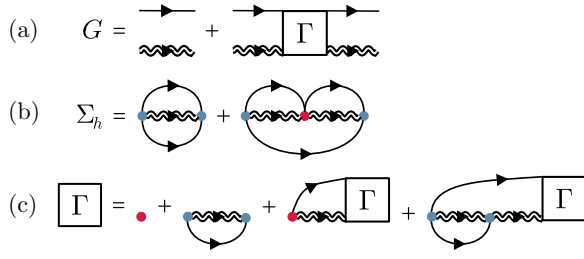


FIG. 2. (a) The Green's function $G(\mathbf{p}, \omega)$ of a physical hole is a two-body Green's function of a spinon and a holon. Solid lines are the spinon propagator and double wavy lines are the dressed holon propagator [Eq. (11)]. (b) Holon self-energy $\Sigma_h(\mathbf{p}, \omega)$ giving the dressing of holons by spinons. (c) Spinon-holon scattering matrix $\Gamma(\mathbf{p}, \mathbf{q}_1, \mathbf{q}_2, \omega)$, with red and blue balls the scattering h and g vertices, respectively, in Eq. (7).

two-body Green's function, shown diagrammatically in Fig. 2(a), is challenging for strong interaction $t_1/J_1 > 1$ due to the spinon-holon interactions given in Eq. (6). These interactions enter in two qualitatively different ways.

First, while the spinons are unaffected by the presence of a single hole so that their propagator is given by $1/(\omega - \omega_{\mathbf{k}}^s)$ in frequency space, the holons can be strongly affected by the dressing with spinons in the QSL. We use a self-consistent diagrammatic approach to include this dressing for the holon propagator, which in frequency space reads

$$G_h(\mathbf{p}, \omega) = \frac{1}{\omega - \omega_{\mathbf{p}}^h - \Sigma_h(\mathbf{p}, \omega)}. \quad (11)$$

This amounts to resumming a class of noncrossing diagrams for the holon self-energy $\Sigma_h(\mathbf{p}, \omega)$ to infinite order as shown in Fig. 2(b) and described in detail in Refs. [46, 47]. Our approach is analogous to the self-consistent Born approximation for hole motion in an antiferromagnet [48], which has proven to be very accurate when compared with experimental and exact numerical results [49–51]. Inclusion of this dressing of the holon propagator but assuming that it moves independently of the spinon corresponds to the first diagram in Fig. 2(a). Such uncorrelated motion is equivalent to the hole fractionalizing into an unbound spinon-holon pair. It gives rise to a broad spectral response of the hole as was analyzed in detail in Ref. [46], and this absence of a well-defined quasiparticle peak has been proposed as a characteristic feature of QSLs [52–54].

A second way of interaction enters when one is calculating the hole Green's function between the holon and the spinon that are simultaneously created when the electrons is removed. Such interactions are described by the holon-spinon scattering matrix Γ entering the second diagram in Fig. 2(a). Given the diagrams we use to calculate the holon Green's function shown in Fig. 2(b), we must include the diagrams generated by the Bethe-Salpeter equation shown

in Fig. 2(c) to obtain a conserving approximation for the two-body holon-spinon Green's function [55]. Doing this to infinite order is, however, very challenging, and in practice we instead numerically iterate the Bethe-Salpeter equation order by order, which is then used to calculate the hole Green's function. As detailed in Appendix B, we find that convergence is quickly obtained after a few iterations for most momenta and that the spinon-holon interactions have little effect on the hole Green's function, which is well-described by the uncorrelated first diagram in Fig. 2(a). This gives a broad spectral response coming from the fractionalization of the electron as expected for a spin liquid. For momenta in the vicinity of $\mathbf{k} = (\pi, \pi)$, we however cannot obtain convergence with the hole spectral function changing significantly from one iteration to the next. We now show that this behavior is due to a singularity arising from the presence of a bound state that cannot be described by finite-order perturbation theory.

IV. EFFECTIVE SCHRÖDINGER EQUATION

We now derive an effective Schrödinger equation for the spinon-holon pair, which automatically includes the spinon-holon scattering events in Γ to infinite order in a much simpler way than solving the full Bethe-Salpeter equation.

To do this, we first notice that diagrams involving the spinon pair creation/annihilation vertex g [blue vertices in Fig. 2(c)] turn out to give small contributions and do not capture the divergences; see Appendix B for details. It is, therefore, sufficient to include the dominant ladder diagrams involving the vertex h [diagrams with red vertices in Fig. 2(c)] to identify any bound states, which describe repeated spinon-holon scatterings to infinite order. This gives the Bethe-Salpeter equation

$$\Gamma(\mathbf{p}, \mathbf{k}, \mathbf{k}', \omega) = h_{\mathbf{p}, \mathbf{k}, \mathbf{k}'} + \sum_{\mathbf{q}} h_{\mathbf{p}, \mathbf{k}, \mathbf{q}} G(\mathbf{p} - \mathbf{q}, \omega - \omega_{\mathbf{k}+\mathbf{q}}^s) \times \Gamma(\mathbf{p} - \mathbf{q}, \mathbf{k} + \mathbf{q}, \mathbf{k}' - \mathbf{q}, \omega). \quad (12)$$

We then use a pole expansion of the holon Green's function, writing

$$G_h(\mathbf{p}, \omega) \simeq \frac{Z_{\mathbf{p}}^h}{\omega - \tilde{\omega}_{\mathbf{p}}^h + i\gamma_{\mathbf{p}}^h}, \quad (13)$$

where $\tilde{\omega}_{\mathbf{p}}^h$ is the energy of a dressed holon with momentum $\mathbf{p}$, determined by solving $\tilde{\omega}_{\mathbf{p}}^h = \omega_{\mathbf{p}}^h + \text{Re } \Sigma_h(\mathbf{p}, \tilde{\omega}_{\mathbf{p}}^h)$ numerically, $Z_{\mathbf{p}}^h$ is its residue, and $\gamma_{\mathbf{p}}^h = -Z_{\mathbf{p}}^h \text{Im } \Sigma_h(\mathbf{p}, \tilde{\omega}_{\mathbf{p}}^h)$ is half its the decay rate [46], which can be nonzero due to spinon-holon interactions.

Finally, we define an effective interaction vertex $\tilde{h}_{\mathbf{p}, \mathbf{k}, \mathbf{k}'} = (Z_{\mathbf{p}}^h)^{1/2} h_{\mathbf{p}, \mathbf{k}, \mathbf{k}'} (Z_{\mathbf{p}-\mathbf{k}'}^h)^{1/2}$ and scattering matrix

$\tilde{\Gamma}(\mathbf{p}, \mathbf{k}, \mathbf{k}', \omega) = (Z_{\mathbf{p}}^h)^{1/2} \Gamma(\mathbf{p}, \mathbf{k}, \mathbf{k}', \omega) (Z_{\mathbf{p}-\mathbf{k}}^h)^{1/2}$ so that Eq. (12) can be written as

$$\begin{aligned} \tilde{\Gamma}(\mathbf{p}, \mathbf{k}, \mathbf{k}', \omega) &= \tilde{h}_{\mathbf{p}, \mathbf{k}, \mathbf{k}'} + \sum_{\mathbf{q}} \\ &\times \frac{\tilde{h}_{\mathbf{p}, \mathbf{k}, \mathbf{q}}}{\omega - \omega_{\mathbf{k}+\mathbf{q}}^s - \tilde{\omega}_{\mathbf{p}-\mathbf{q}}^h + i\gamma_{\mathbf{p}-\mathbf{q}}^h} \tilde{\Gamma}(\mathbf{p} - \mathbf{q}, \mathbf{k} + \mathbf{q}, \mathbf{k}' - \mathbf{q}, \omega). \end{aligned} \quad (14)$$

With Eq. (14), we have reduced the complicated Bethe-Salpeter equation to a much simpler Lippmann-Schwinger equation describing the two-body scattering of a spinon-holon pair. This in turn is equivalent to a Schrödinger equation $\hat{H}_{\text{eff}}|\Psi\rangle = E|\Psi\rangle$ for the relative wave function $|\Psi\rangle$ of the spinon-holon pair with the effective Hamiltonian [56]

$$\begin{aligned} \hat{H}_{\text{eff}} &= \sum_{\mathbf{k}} (\tilde{\omega}_{\mathbf{k}}^h - i\gamma_{\mathbf{k}}^h) \tilde{b}_{\mathbf{k}}^{\dagger} \tilde{b}_{\mathbf{k}} + \sum_{\mathbf{k}} \omega_{\mathbf{k}}^s \hat{\gamma}_{\mathbf{k}}^{\dagger} \hat{\gamma}_{\mathbf{k}} \\ &+ \sum_{\mathbf{k}, \mathbf{k}'} \tilde{h}_{\mathbf{p}-\mathbf{k}, \mathbf{k}, \mathbf{k}'-\mathbf{k}} \tilde{b}_{\mathbf{p}-\mathbf{k}'}^{\dagger} \hat{\gamma}_{\mathbf{k}', \sigma}^{\dagger} \tilde{b}_{\mathbf{p}-\mathbf{k}} \hat{\gamma}_{\mathbf{k}, \sigma}. \end{aligned} \quad (15)$$

Here, $\tilde{b}_{\mathbf{k}}^{\dagger}$ creates a dressed holon quasiparticle with momentum $\mathbf{k}$, energy $\tilde{\omega}_{\mathbf{k}}^h$, and decay rate $\gamma_{\mathbf{k}}^h$. Equation (15) provides a general and physically intuitive description of hole dynamics in the QSL in terms of dressed holons interacting with spinons, which is much easier to analyze than the full field theory. Nontrivial many-body effects of the background QSL are included in several ways. First, the dispersion $\omega_{\mathbf{k}}^s$ of the spinons is determined by the background QSL and the dispersion $\tilde{\omega}_{\mathbf{k}}^h$ of the holons includes the dressing by spinons. Second, the spinon-holon interaction is renormalized by the holon residues in analogy with the microscopic foundation of Fermi liquid theory [57,58]. It also depends on all momenta, reflecting the lack of Galilean invariance, making it nonlocal, which is typical for effective Schrödinger equations for many-body systems. Finally, the dressed holons can be damped due to their interactions with the spinons, and Eq. (15) in this sense describes an open system. It was shown in Ref. [46] that dressed holons have a vanishing decay rate around the momentum $\mathbf{p} = (\pi, \pi)$, whereas they become strongly damped and eventually disappear in a region around $\mathbf{p} = (0, 0)$. We determine the energy of these strongly damped holons from their spectral function peak, but since they have a small or vanishing residue they essentially do not interact with any other states and thus have little influence on the dynamics.

Since we have used a number of approximations to arrive at Eq. (15), a natural question is the accuracy of this approach. Here it should be noted that we have used a similar mapping of a Bethe-Salpeter equation to an effective Schrödinger equation to analyze the bound states

between two mobile impurities in a Bose-Einstein condensate. This turned out to be remarkably accurate when compared with Monte-Carlo calculations even for strong interactions [59,60].

V. BOUND STATES AND EMERGING FERMIONS

We now use the effective Hamiltonian given by Eq. (15) to analyze the two-body spinon-holon problem created when an electron is removed from the QSL. In particular, we show that the divergences described in Sec. III when iterating the second diagram in Fig. 2(a) are due to the presence of spinon-holon bound states.

To do this, we solve the Schrödinger equation

$$\hat{H}_{\text{eff}}|\Psi_{\mathbf{p}}^n\rangle = E_{\mathbf{p}}^n|\Psi_{\mathbf{p}}^n\rangle, \quad (16)$$

with

$$|\Psi_{\mathbf{p}}^n\rangle = \sum_{\mathbf{k}} \phi_{\mathbf{p}, \mathbf{k}}^n \tilde{b}_{\mathbf{p}-\mathbf{k}}^{\dagger} \hat{\gamma}_{\mathbf{k}, \downarrow}^{\dagger} |\text{QSL}\rangle \quad (17)$$

the n th two-body eigenstate with COM momentum $\mathbf{p}$. Here, $\phi_{\mathbf{p}, \mathbf{k}}^n$ is the relative spinon-holon wave function and $|\text{QSL}\rangle$ is the QSL ground state at half-filling with $\hat{b}_{\mathbf{p}}|\text{QSL}\rangle = \hat{\gamma}_{\mathbf{k}, \sigma}|\text{QSL}\rangle = 0$. Solving Eq. (16) numerically is much easier than solving the full Bethe-Salpeter equation.

In Fig. 3(a), we plot the energy spectrum of the spinon-holon states $|\Psi_{\mathbf{p}}^n\rangle$ as a function of their COM along straight lines in the BZ for the interaction strength $J_1/t_1 = 0.6$. The spectrum is determined by our taking the real part of the complex energy obtained by solving Eq. (16) numerically. We see that the solutions fall into two distinct classes. First, there is a continuum coming from delocalized scattering states dominated by a single term in the superposition in Eq. (17). These states are the unbound spinon-holon pairs, i.e., the fractionalized fermions expected for a QSL. Notably, we also find three discrete energy bands well below the continuum as highlighted with green, yellow, and red lines. These states, moreover, have a small decay rate on the order of $\sim 10^{-2}$ times the energy gap to the continuum, making them well-defined *bound states*. The reason for their long lifetime is that the bound states are essentially uncoupled from the strongly damped but also weakly interacting holons; see Appendix A for details.

We plot in Fig. 3(b) the wave functions $\phi_{\mathbf{p}, \mathbf{k}}^n$ for bound states at the COM momentum $\mathbf{p} = (\pi, \pi)$. At this momentum, the green band is degenerate with the yellow band, whereas the red band has a slightly higher energy. Figure 3(c) shows the wave function squared in real space obtained by Fourier transform. These plots clearly show that the relative wave function is spread out in momentum space and localized in real space, reflecting that the spinon and holon are bound together. We also see that the wave function of the two lowest degenerate states has a

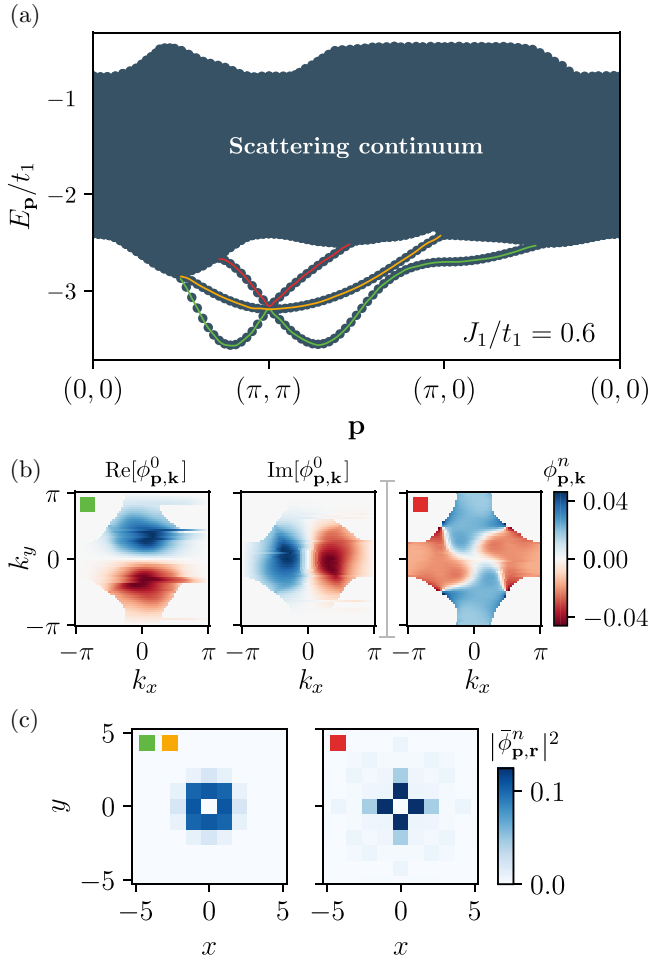


FIG. 3. (a) Spinon-holon energy spectrum obtained from Eqs. (16)–(17) for a 64×64 lattice along straight paths in the Brillouin zone. The red, yellow, and green bands are bound states. (b) Bound state wave functions for $\mathbf{p} = (\pi, \pi)$ in momentum space. The two leftmost panels illustrate the real and imaginary parts of one of the degenerate p -wave states. For the other p -wave state, the imaginary part has changed sign, while the real part is identical. (c) Plots of the gauge-averaged relative wave function in real space, with the left panel showing the p -wave states. This shows that the spin and charge of the quasiparticles are located within a few lattice sites.

p -wave symmetry, whereas the highest energy bound state wave function has d -wave symmetry. Note that the wave functions in momentum space do not have the mirror symmetries of the lattice due to our specific choice of phase (gauge) of the mean-field $\Delta_{\mathbf{k}}$ in Eq. (4) describing the singlets of the QSL. We have averaged over this gauge for the real-space plot in Fig. 3(c), thereby recovering these symmetries. This accounts for the different symmetry-broken states realized in repeated experiments. It should be noted that at half-filling, these symmetries are naturally present due to an $SU(2)$ gauge [61]; see Appendix D for details.

From this, we conclude that the QSL for certain COM momenta supports bound spinon-holon states with

energies below the usual scattering continuum, a long lifetime, and an internal spatial structure extending over a few lattice sites. These states, therefore, have a well-defined energy, spin, and charge, and they correspond to new electronlike fermions. We emphasize that these fermions emerge from our theory based on the single-band Hubbard model in the strong-coupling limit with no need to add further degrees of freedom, and that they have no obvious smooth connection to the electronic quasiparticles present at weak interactions. This is a main result of the present paper and it provides a microscopic mechanism for the conjecture that the pseudogap phase of the cuprates consists of a so-called FL* existing on top of a QSL [21–30]. We explore the FL* phase existing for a finite concentration of holes further in Sec. VI B. In Appendix C, we show by tuning the pairing potential that the formation of these quasiparticles is stabilized by shorter singlets in the QSL.

VI. OBSERVABLES

We now turn to how these emerging fermions and the FL* can be observed in condensed matter as well as optical lattice systems. Since the fermions are bound states of spinons and holons with a very long lifetime from the low-energy part of the spectrum, we ignore the holon decay in the following. In Appendix A it is shown that this is a very good approximation.

A. Spatial structure

Using quantum microscopy with atoms in optical lattices, one can take pictures of many-body systems with single-site resolution [62]. In particular, the spin correlations have been measured in the neighborhood of mobile holes (or particles) added to a Fermi Hubbard system at half-filling [10,11,63–65].

In the parton construction, a vacant site means it is occupied by a holon, and we, therefore, analyze the correlation function

$$M_{\mathbf{p}}^n(\mathbf{d}) = \frac{\langle \Psi_{\mathbf{p}}^n | \hat{S}_{\mathbf{r}+\mathbf{d}}^z \hat{b}_{\mathbf{r}}^{\dagger} \hat{b}_{\mathbf{r}} | \Psi_{\mathbf{p}}^n \rangle}{\langle \Psi_{\mathbf{p}}^n | \hat{b}_{\mathbf{r}}^{\dagger} \hat{b}_{\mathbf{r}} | \Psi_{\mathbf{p}}^n \rangle} = -\frac{1}{2N} \left(\left| \sum_{\mathbf{k}} \phi_{\mathbf{p},\mathbf{k}}^n u_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{d}} \right|^2 + \left| \sum_{\mathbf{k}} \phi_{\mathbf{p},\mathbf{k}}^n v_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{d}} \right|^2 \right) \quad (18)$$

for the spin $\hat{S}_{\mathbf{r}+\mathbf{d}}^z = (\hat{f}_{\mathbf{r}+\mathbf{d},\uparrow}^{\dagger} \hat{f}_{\mathbf{r}+\mathbf{d},\uparrow} - \hat{f}_{\mathbf{r}+\mathbf{d},\downarrow}^{\dagger} \hat{f}_{\mathbf{r}+\mathbf{d},\downarrow})/2$ observed at $\mathbf{r} + \mathbf{d}$ given that the holon is observed at $\mathbf{r}$, assuming that the system is in the quantum state $|\Psi_{\mathbf{p}}^n\rangle$. The normalization gives $\langle \Psi_{\mathbf{p}}^n | \hat{b}_{\mathbf{r}}^{\dagger} \hat{b}_{\mathbf{r}} | \Psi_{\mathbf{p}}^n \rangle = 1/N$ since the holon is uniformly distributed in the lattice. We also have $\sum_{\mathbf{d}} M_{\mathbf{p}}^n(\mathbf{d}) = -1/2$, reflecting that a single spin-up electron has been removed from the half-filled QSL.

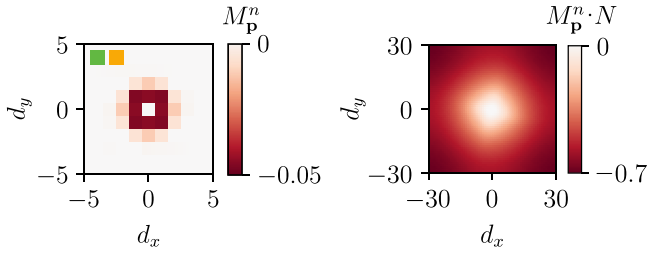


FIG. 4. Spin-hole correlation function $M_{\mathbf{p}}^n(\mathbf{d})$ given by Eq. (18) for $\mathbf{p} = (\pi, \pi)$, $J_1/t_1 = 0.6$, and a 64×64 lattice. The left panel shows the p -wave bound state and the right panel shows a delocalized state in the scattering continuum. Note that the scale in the right panel is multiplied by N .

The left panel in Fig. 4 shows the spin-hole correlation function $M_{\mathbf{p}}^n(\mathbf{d})$ for the lowest bound state $|\Psi_{\mathbf{p}}^n\rangle$ with momentum $\mathbf{p} = (\pi, \pi)$. We have again performed a gauge averaging to simulate the shot-to-shot variations in an experiment; see Appendix D. This clearly shows that the net spin (magnetization) is spatially localized in the vicinity of the hole, reflecting that the spinon and holon have formed a fermionic quasiparticle with well-defined charge and spin localized over a few lattice sites. The symmetry of the magnetization around the hole is directly inherited from the relative spinon-holon wave function shown in Fig. 3(c). In contrast, the right panel in Fig. 4 shows $M_{\mathbf{p}}^n(\mathbf{d})$ for one of the scattering states. Here, the magnetization is of order $1/N$ on each lattice site and is distributed over the whole lattice even when the hole is observed at a give lattice site. This explicitly shows the fractionalization of the fermions into unbound charge (holon) and spin (spinon) excitations characteristic of QSLs [20].

Note that we omitted the dressing of the holons by spinons as described by Σ_h when deriving the second line in Eq. (18). This dressing cloud would also frustrate the singlet environment surrounding the holon, but this dressing is $SU(2)$ symmetric, so $\langle \hat{S}_{\mathbf{r}}^z \rangle = 0$ everywhere for an isolated dressed holon, reflecting that it carries charge but no spin. This makes it possible for $M_{\mathbf{p}}^n(\mathbf{d})$ to distinguish between bound spinon-holon pairs and uncorrelated quasiparticles.

These results, therefore, show that optical lattices are a promising platform for observing the internal spatial structure of the emerging fermions in the FL*, provided that the relevant parameter and temperature regimes of the Hubbard model can be realized. The spin correlation function $M_{\mathbf{p}}^n(\mathbf{d})$ is *simpler* than those already measured around holes in a Fermi-Hubbard antiferromagnet, which involve at least two spin operators [10,11,63–65].

B. Spectral properties

ARPES is a very successful technique for probing the spectral properties of condensed-matter systems. It has, in particular, been used to observe the characteristic “Fermi

arcs” in the pseudogap phase of unconventional superconductors [66–68], whose microscopic origin remains an open and important question [30]. The spectral properties of magnons in optical lattices have furthermore recently been measured by radio-frequency spectroscopy [69], and this technique can be used to probe the quasiparticles formed by mobile holes in a Fermi-Hubbard system [70].

We, therefore, analyze the spectral properties of the FL* as probed by these techniques. The contribution to the hole spectral function $A(\mathbf{p}, \omega) = -2 \text{Im} G(\mathbf{p}, \omega)$ coming from the uncorrelated (fractionalized) spinon-holon pairs was analyzed in Ref. [46] by evaluation of the first diagram in Fig. 2(a). Here, we focus on spinon-holon bound states as described by the second diagram in Fig. 2(a). With use of the Lehman representation and Eq. (17), this can be written as

$$A(\mathbf{p}, \omega) = \frac{2}{N} \sum_n \frac{\eta}{(\omega - E_{\mathbf{p}}^n)^2 + \eta^2} \left| \sum_{\mathbf{k}} v_{\mathbf{k}} \phi_{\mathbf{p}, \mathbf{k}}^n \right|^2, \quad (19)$$

where we have added the generic small imaginary part, $\eta = 0.01t_1$, also used when the analytical continuation for the holon’s Green’s function is performed. In the following plots, Figs. 5 and 6, we have again performed a gauge averaging of the spectral function.

Consider first the spectral function for the high-symmetry momentum $\mathbf{p} = (\pi, \pi)$ shown in Fig. 5(a). The vertical dashed lines indicate the energies $E_{\mathbf{p}}^n$ of the bound states shown in Fig. 3. We see that the highest bound state gives rise to a clear quasiparticle peak in the spectrum, whereas the two lowest states are *not* visible, i.e., they are

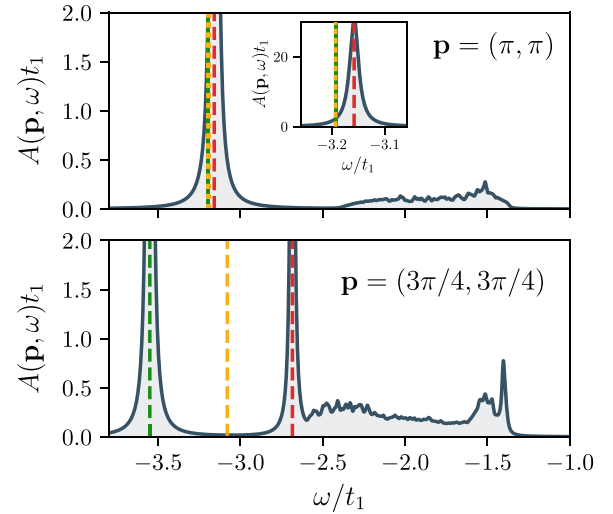


FIG. 5. Hole spectral function given by Eq. (19) for two different momenta. The vertical dashed lines shows the energy of the bound states with the same color code as in Fig. 3. The inset shows an enlarged section around the peak.

“dark.” This is due to the sum $\sum_{\mathbf{k}} \phi_{\mathbf{p},\mathbf{k}}^n v_{\mathbf{k}}$ in Eq. (19), giving the overlap between the spinon-holon wave function and the state $\hat{c}_{\mathbf{p},\uparrow} |\text{QSL}\rangle = \sum_{\mathbf{k}} v_{\mathbf{k}} \hat{b}_{\mathbf{p}-\mathbf{k}}^\dagger \hat{\gamma}_{\mathbf{k},\downarrow}^\dagger |\text{QSL}\rangle / N$ created by removal of an electron from the QSL. The state created by removal of an electron, $\hat{c}_{\mathbf{p},\uparrow} |\text{QSL}\rangle$, inherits the d_{xy} -wave symmetry of the QSL given by Eq. (4), which makes its overlap with the two lowest states at $\mathbf{p} = (\pi, \pi)$ vanish because they have p -wave symmetry. The overlap, on the other hand, is nonzero for the third state since it has d -wave symmetry. In Fig. 5(b), the spectral function is plotted for $\mathbf{p} = (3\pi/4, 3\pi/4)$ where the bound states have a lower symmetry. They are consequently all bright in the hole spectral function, although the spectral weight of the middle (yellow) energy state is very small because the contribution from the d_{xy} -wave pairing channel is small.

To further illustrate how the interplay between the internal structure of the fermions and the symmetry of the singlet in the QSL show up in the spectral properties, we plot in Fig. 6(a) the spectral function $A(\mathbf{p}, \omega)$ along straight lines in the BZ. This shows that the yellow (middle) band in Fig. 3 has a very low spectral weight, making it almost dark. We also see that the spectral weight of the lowest band decreases when we move from the minimum around $\mathbf{p} = (3\pi/4, 3\pi/4)$ toward the high-symmetry point $\mathbf{p} = (\pi, \pi)$, where the interaction wave function becomes p -wave symmetric, making it dark as discussed above.

These results demonstrate that the emerging fermions can be detected in ARPES as peaks in the hole spectral function with a spectral weight determined by how their internal spatial symmetry relates to the symmetry of the QSL state.

C. Fractionalized Fermi liquid and Fermi arcs

In analogy with the weakly interacting Feshbach dimers formed by strongly bound atoms [71], we expect the emerging fermions to be weakly interacting when their binding energy is large so that their spatial size is a few lattice sites. For a nonzero hole concentration x , they will then form a so-called FL*. In the following, we focus on the low-doping regime, where spin liquids have been found to remain stable, and examine how this manifests itself in ARPES experiments [72,73].

Figures 6(b) and 6(c) show the spectral function in the BZ for different Fermi energies ε_F of the FL* corresponding to the hole-filling fractions $x = 0.01$, $x = 0.02$, and $x = 0.04$, and for different values of J_1/t_1 ; see also Fig. 1. That is, ε_F is the highest energy of the occupied spinon-holon bound states for given filling fraction x . We see that the holes form anisotropic pockets located at the corners of the BZ with a low-intensity back side. Comparing Figs. 6(b) and 6(c), we see that the arc features are robust with respect to doping and interaction strength, whereas their exact shape and size depend quite sensitively on these parameters. Again, this suppression of intensity is a

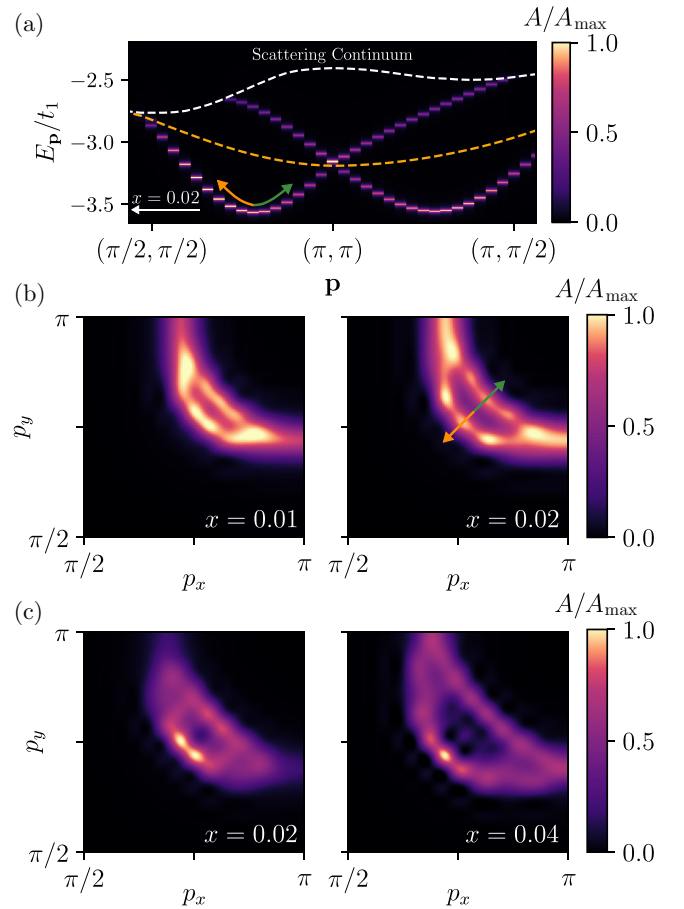


FIG. 6. (a) Spectral function $A(\mathbf{p}, \omega)$ along straight paths in the Brillouin zone for $J_1/t_1 = 0.6$. The dashed yellow line indicates the position of the yellow (middle) band in Fig. 3. The white arrow indicates the Fermi energy for the filling fraction $x = 0.02$ used in (b). Spectral function at the Fermi surface of the FL* for the indicated hole doping x and for (b) $J_1/t_1 = 0.6$ and (c) $J_1/t_1 = 0.3$. These clearly display hole pockets with dimly lit back sides. There are three identical hole pockets in the other corners of the Brillouin zone. The orange and green arrows in (b) indicate the same directions in the Brillouin zone as in (a).

result of these bound states predominately having p -wave symmetry, giving a suppressed overlap with the d -wave singlets. The green and orange arrows in Fig. 6(b) indicate the paths in the BZ in Fig. 6(a), where the Fermi level for $x = 0.02$ is also shown. Note that the area of the hole pockets in Figs. 6(b) and 6(c) is given by their concentration x , which is quite different from the area $1 + x$ of weakly interacting holes smoothly connected to the noninteracting holes. Luttinger’s theorem [74,75] is, therefore, broken by the FL*, showing that it is distinct from the Fermi liquid present at weak interactions [21,23].

Remarkably, the hole pockets in Fig. 6 are qualitatively similar to the Fermi arcs measured with ARPES in the pseudogap phase of the cuprates [66–68,76,77], with respect to both their shape and their anisotropic spectral

weight. Recovering these features is highly nontrivial since our microscopic theory is based on a single-band t - J model from which the FL* emerges for strong interactions as a Fermi liquid of bound spinon-holon states with no extra ingredients required. The predicted Fermi arcs are a result of the delicate interplay between the energies of these fermions as well as their internal spatial structure. Our results, therefore, provide a microscopic mechanism for the conjecture that the pseudogap phase is a FL* coexisting with a QSL background, which has been used to explain quantum oscillation and transport measurements, as well as the observed Fermi arcs [20,22–28,30]. To our knowledge, most calculations supporting this conjecture have so far been based on phenomenological descriptions or models where the fermions effectively are added on top of the background QSL on the basis of physical arguments.

VII. DISCUSSION AND OUTLOOK

Using a field-theoretic approach, we explored the dynamics of holes in a background $\mathbb{Z}_2$ QSL as described by a single-band, extended t - J model close to half-filling. The spinon-holon scattering was shown to diverge for certain COM, and we then derived an effective low-energy Hamiltonian to demonstrate that this divergence is due to spinon-holon bound states. These bound states correspond to the existence of well-defined fermions with the same charge and spin as the underlying electrons, which is unexpected from the general paradigm of uncorrelated spin and charge degrees of freedom in QSLs. A nonzero concentration of holes is then shown to lead to hole pockets whose ARPES spectra exhibit the qualitative features of the Fermi arcs observed in the cuprate pseudogap phase with respect to both the shape and the position in the Brillouin zone.

Since the shape and anisotropy of the Fermi-surface emerges from our theory based on a single-band t - J model due to the interplay between the symmetries of the spin liquid and the internal structure of the fermions with no further degrees of freedom needed, we in this sense provide microscopic support for the conjectured existence of a FL* in doped spin liquids, which may in turn be the origin of the pseudogap phase observed in the cuprates [21–30]. We additionally expect that the Fermi surface of a FL* is highly anisotropic, unless the pocket is located at a high-symmetry point, because the intensity of the ARPES signal is determined by this subtle interplay.

In particular, our theory gives a precise mechanism for the binding of holons and spinons into new fermions, which forms the basis for a number of phenomenological theories, such as quantum dimer models [24,27,28]. It would in this connection be interesting to explore if and how the emerging fermions in our theory are connected to the “ancilla” model, where electrons are coupled to qubits in two layers added as a mathematical tool to model strong

electron correlations [29,30,78]. Quantum gas microscopy with optical lattices makes it possible to probe the real-space properties of electronlike quasiparticles in the t - J model via the spin-hole correlation function, as has already been done for antiferromagnets [10,11,63–65]. Here, it should be noted that the temperatures required to reach the pseudogap phase are significantly higher than those required to achieve superconductivity.

Our theoretical framework opens up new ways to explore hole motion in quantum spin liquids and its possible relation to pseudogap physics. It is, therefore, interesting to explore the properties of the emerging fermions for different values of the parameters entering the t - J model. Also, even though it is often argued that a single-band model captures the essential physics of the cuprates, they are clearly very complex, and an important future research direction is, therefore, to extend the t - J model to include, for example, several bands. Exploring possible Cooper pairing instabilities of the emerging fermions and whether they occur in the d -wave channel would provide strong indications that the FL* is indeed a parent state of high- T_c superconductors. We note that recent density-matrix renormalization group (DMRG) calculations strongly indicate an instability toward pairing for the same extended t - J model as considered here within the same parameter range, using cylinders of up to six-site circumference [79]. A pronounced asymmetry between electron-doped and hole-doped systems was predicted in these calculations, and valuable insights would, therefore, be obtained by performing a similar analysis within our framework with a positive $t_2/t_1 = +\sqrt{J_2/J_1}$ corresponding to electron doping. Another question concerns if our theoretical framework predicts a quantum phase transition from the FL* to a conventional Fermi liquid at a critical hole concentration as observed experimentally [80]. One should also explore if the lifetime and in-plane conductivity of the emerging fermions exhibit Fermi-liquid-like behavior as has been observed in optical conductivity and magnetoresistance measurements [81,82].

Since the pseudogap phase seems to be a universal phenomenon appearing in many different materials, a key problem concerns whether the emerging fermions are a robust feature of mobile holes in QSLs. More broadly, it is interesting to investigate whether such fermions exist in other kinds of lattice geometry and QSL. This naturally raises the question of the effects of gauge fluctuations. While they likely are not important for the $\mathbb{Z}_2$ QSL studied here due to their gapped nature making the low-energy spectrum of the mean-field description similar to that found in variational Monte Carlo studies [44], gauge fluctuations may be important for, e.g., U(1) QSLs in triangular lattices [61,83,84]. With this in mind, it would be very useful to compare results from our theory with those from numerical methods such as matrix-product-state-based dynamical algorithms [53,54] or the variational

Monte Carlo method [85]. Additionally, as spin liquid phases become unstable with increasing doping [72,86,87], it would be informative to determine the pairing fields $\Delta_{\mathbf{k}}$ as a function of doping, since the bound state formation and spectral properties are dependent on these; see Appendix C. Finally, DMRG studies suggest the use of a broad spectral response arising from the fractionalization of electrons into uncorrelated holons and spinons as a signature of a QSL [52–54]. Our results, on the other hand, show that this is not always the case, and one should, therefore, explore how common the emerging fermions are.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data supporting this study's findings are available within the article.

APPENDIX A: OPEN SYSTEM DYNAMICS

In this appendix we further examine the consequences of considering the spinon and holon to be interacting in an open system where the holon quasiparticle can decay. To understand and perform calculations for this open system, we follow the approach described in Ref. [88]. We compare these results with calculations where the decay of the holon quasiparticle is omitted.

Solving the effective Schrödinger, we retrieve complex energies where the real part reflects the observable energy of the state and the imaginary part reflects decay. In Fig. 7(a), we compare the bottom of the energy spectrum when considering decay (blue balls) and when decay is ignored (red circles). This shows no visible difference, and we find the average difference between the two ways of calculating the energies to be $\sim 10^{-4} \cdot t_1$.

Including the decay of the holons means that the decay of the spinon-holon states is given by the imaginary part of the energy. Using the framework developed in Ref. [88], we define the right and left states as $\hat{H} |\Psi_{\mathbf{p},r}^n\rangle = E_{\mathbf{p}}^n |\Psi_{\mathbf{p},r}^n\rangle$

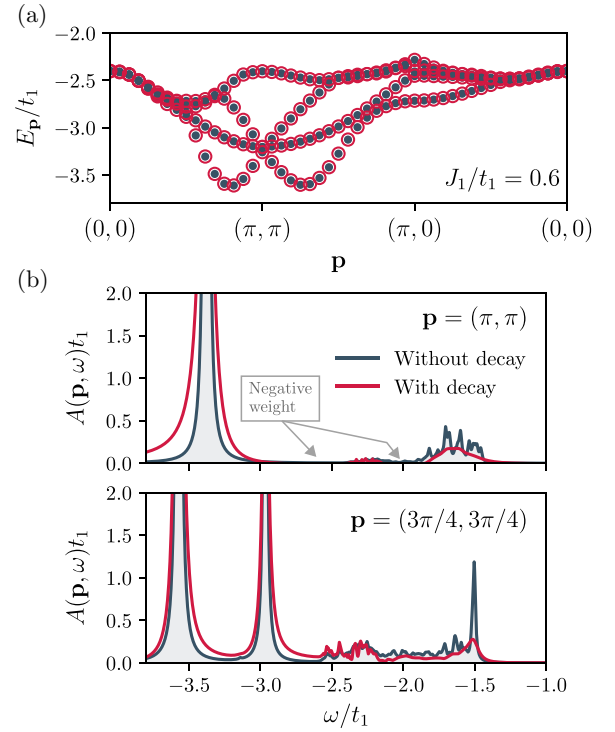


FIG. 7. (a) The four lowest energies found by solving the effective Hamiltonian [Eq. (16)] with decay (red circles) and without decay (blue balls). Calculated for a system size of 32×32 , (b) shows the spectral function calculated with Eq. (A2) in red and calculated with Eq. (19) in blue for the indicated momenta. Gray arrows indicate areas of negative spectral weight when decay is considered.

and $\langle \Psi_{\mathbf{p},l}^n | \hat{H} = E_{\mathbf{p}}^n \langle \Psi_{\mathbf{p},l}^n |$, respectively. This leads to

$$\begin{aligned} |\Psi_{\mathbf{p},r}^n\rangle &= \sum_{\mathbf{k}} \phi_{\mathbf{p},\mathbf{k}}^n \tilde{b}_{\mathbf{p}-\mathbf{k}}^\dagger \hat{\gamma}_{\mathbf{k},\downarrow}^\dagger |\text{QSL}\rangle, \\ |\Psi_{\mathbf{p},l}^n\rangle &= \sum_{\mathbf{k}} [\phi_{\mathbf{p},\mathbf{k}}^n]^* \tilde{b}_{\mathbf{p}-\mathbf{k}}^\dagger \hat{\gamma}_{\mathbf{k},\downarrow}^\dagger |\text{QSL}\rangle, \end{aligned} \quad (\text{A1})$$

and $\sum_{\mathbf{p},n} |\Psi_{\mathbf{p},r}^n\rangle \langle \Psi_{\mathbf{p},l}^n| = \mathbb{1}$. With use of the Lehmann representation of the Green's function the spectral function reads

$$A(\mathbf{p}, \omega) = -\frac{2}{N} \sum_n \text{Im} \left[\frac{1}{\omega - E_{\mathbf{p}}^n} \left(\sum_{\mathbf{k}} v_{\mathbf{k}} \phi_{\mathbf{p},\mathbf{k}}^n \right)^2 \right], \quad (\text{A2})$$

when decay is considered. In Fig. 7(b), we see the spectral function calculated with Eq. (A2) in red. In blue, we see the spectral function calculated with Eq. (19). This again shows that including the decay does not change the behavior at the bottom of the spectrum. Considering decay, we also see that the spectral function can take negative values in the scattering continuum. This is not uncommon when considering a non-Hermitian Hamiltonian and illustrates

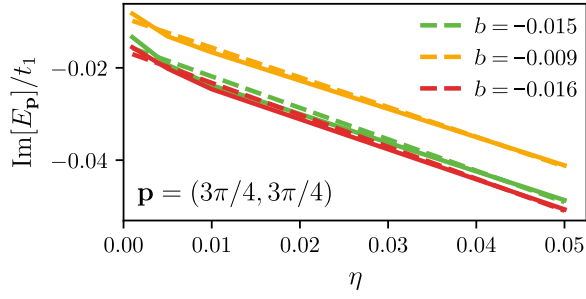


FIG. 8. As a function of the artificial broadening η , we plot the imaginary part of the energy for the three bound states. With dashed lines, we plot the linear fit $a \cdot \eta + b$. The color code is the same as used in Fig. 3.

that these states consist of a superposition that includes holon states with a large decay.

We also find that the bound states are broadening by including decay, which comes from the states having a finite overlap with holon states with a finite decay. In Fig. 8, we investigate the decay of the bound state, $\text{Im}[E_p]$, for $\mathbf{p} = (3\pi/4, 3\pi/4)$ as a function of the infinitesimal broadening η . Here, we see that it converges to a finite value as $\eta \rightarrow 0$. This value is $\sim 10^{-2} \cdot t_1$, which for the ground state is a factor of 10^2 larger than the energy gap to the scattering continuum, making this quasiparticle state well-defined.

APPENDIX B: ITERATIVE APPROACH TO CALCULATING THE GREEN'S FUNCTION OF THE HOLE

In this appendix we calculate the Green's function of the hole, Eq. (10), iteratively by setting up a Bethe-Salpeter equation for the scattering matrix. We first expand the Green's function in terms of Feynman diagrams as

$$G(\mathbf{p}, \omega) = \text{diagram 1} + \text{diagram 2} + \text{diagram 3} + \text{diagram 4} + \dots \quad (\text{B1})$$

with the red vertex corresponding to the h vertex and the blue vertex corresponding to the g vertex in Eq. (6). With $J_1/J_2 = t_1^2/t_2^2 = 0.5$ all the vertices become proportional to t_1/J_1 , leaving no small parameter to expand in. We initiate these calculations by calculating the holon's Green's function, the full wavy line in Eq. (B1), as done in Ref. [46]. Wanting to capture nonperturbative results, we then set up a Bethe-Salpeter equation for the scattering matrix defined in Fig. 2(c). This means we include a subset of the all diagrams, but we include these in a self-consistent way, and in such a way that we retrieve a conserving approximation according to Refs. [55,89]. Now, by calculating the

scattering matrix Γ iteratively until convergence, we can calculate the Green's function by inserting this into the expression in Fig. 2(a).

Before performing the full calculations, let us first understand the impact of the two different types of diagram considered. To do this, we separate the scattering matrix into

$$\begin{aligned} \Gamma_1(\mathbf{p}, \mathbf{q}_1, \mathbf{q}_2, \omega) &= \boxed{\Gamma_1} \\ &= + \text{diagram 1} + \text{diagram 2} + \text{diagram 3} + \dots \\ &= + \text{diagram 4} + \boxed{\Gamma_1} \end{aligned} \quad (\text{B2})$$

and

$$\begin{aligned} \Gamma_2(\mathbf{p}, \mathbf{q}_1, \mathbf{q}_2, \omega) &= \boxed{\Gamma_2} \\ &= \text{diagram 5} + \text{diagram 6} + \dots \\ &= \text{diagram 7} + \boxed{\Gamma_2} \end{aligned} \quad (\text{B3})$$

which leaves out the diagrams mixing the two types of diagram but allows an investigation of the individual contributions. By setting $\Gamma = \Gamma_2$ in the expression for the Green's function, we find for $J_1/t_1 = 0.6$ the results presented in Fig. 9. In green, we also present the first term in Eq. (B1), which is the noninteracting co-propagating term. Now, while these calculations converge and fulfill the sum rule with deviations of less than 1%, we see that

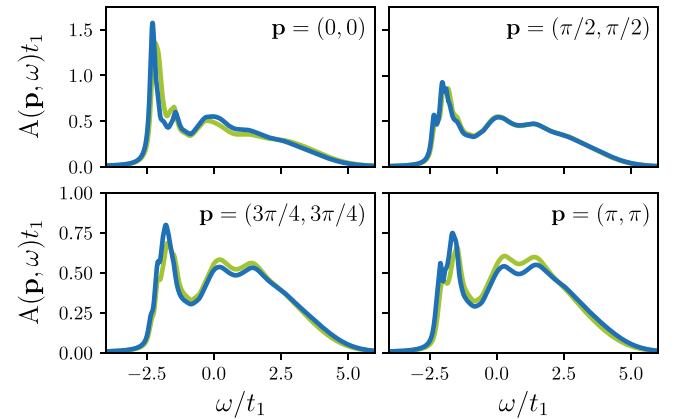


FIG. 9. For a system size of 8×8 , in blue we plot the hole spectral function obtained by use of Eq. (B3) as the scattering matrix. Here, $J_1/t_1 = 0.6$, $\eta = 0.1$, and the crystal momenta are indicated in the respective panels. In green, we plot the spectral function when only considering the co-propagating term.

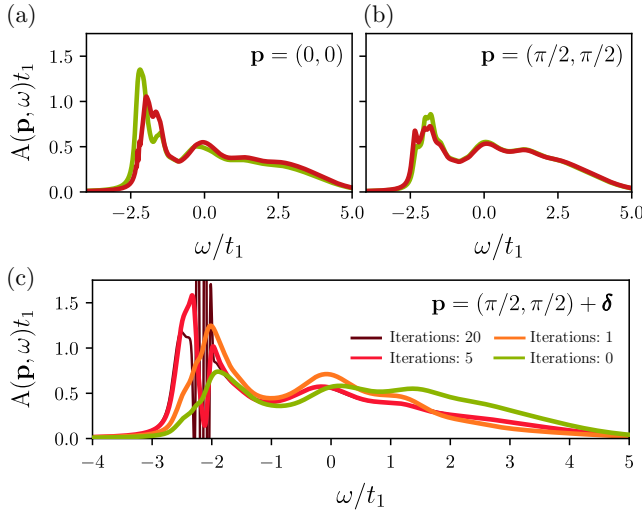


FIG. 10. In red, we plot the hole spectral function computed with Eq. (B3) as the scattering matrix. For comparison with the results from Fig. 9, we use a system size of 8×8 for (a),(b) and use a system size of 16×16 for (c). $J_1/t_1 = 0.6$, $\eta = 0.1$, and the crystal momenta are indicated in the respective panels with $\delta = (2\pi/L, 2\pi/L)$. In green, we plot the spectral function when considering only the co-propagating term. The calculation does not converge in (c) we, therefore, plot several calculations terminated after a different number of iterations.

including the interactions barely changes the spectral function. This indicates that most of the dynamics is governed by the co-propagating term, meaning that the holon and the spinon do not bind and that spin-charge separation is a good assumption. Similar results were found for different interaction strengths and crystal momenta.

Instead, choosing $\Gamma = \Gamma_1$, we obtain the spectral function presented in Fig. 10. For the plots at the top, we again find that the co-propagating term dominates and that the interactions do not change much. In contrast, the lower panel shows that the interactions move spectral weight to the bottom of the spectrum, but ultimately diverge. For each additional iteration the frequency and amplitude of the oscillations increase. We find similar divergences for crystal momenta in the vicinity of $\mathbf{p} = (\pi, \pi)$. Finally, including the full scattering matrix from Fig. 2(c), we see in Fig. 11 that the scattering matrix from Eq. (B2) captures the same divergences, and hence the essential dynamics of these nonperturbative results.

The conclusion from this analysis is that away from $\mathbf{p} = (\pi, \pi)$, the Green's function is well-approximated by the co-propagating term. Close to $\mathbf{p} = (\pi, \pi)$, we see that the type of interactions considered in Eq. (B3) is irrelevant, and instead, the scattering interactions considered in Eq. (B3) are relevant. By considering these, we find indications of nonperturbative features occurring around $\mathbf{p} = (\pi, \pi)$. These features come in the form of the divergences, and we believe these stem from the approach

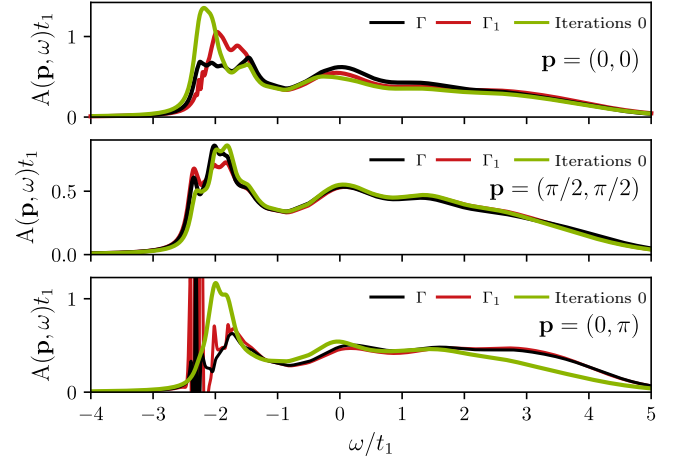


FIG. 11. Spectral function calculated with the full scattering matrix from Fig. 2(c) (black), with Eq. (B2) (red), and with the co-propagating term (green). These calculations are performed for a system size of 8×8 and for $J_1/t_1 = 0.6$, $\eta = 0.1$, and the indicated momenta.

trying to perturbatively capture a pole, hence a spinon-holon bound state. Additional support for this conclusion is found in the Sec. V of the main text, where the analysis predicts bound states at the crystal momenta where the divergences occur.

APPENDIX C: LENGTH OF SINGLETs

One of the motivations for using a BCS-type Hamiltonian to describe the resonating valence bond (RVB) state is that when the Gutzwiller operator is applied on the BCS wave function, the Cooper pairs become resonating singlets and we obtain an RVB state [90]. The pairing potential, therefore, determines the nature of the singlet pairings. In this appendix, we explore the results of having a RVB background with longer singlets. We do this by decreasing Δ_1/ϵ while keeping Δ_1/Δ_2 constant in Eq. (3). In doing this, we soften the spinon spectrum and change the average length of the singlets as described in Ref. [46].

In Fig. 12(a) we see the energy spectrum calculated with $\Delta_1/\epsilon = 1.08$, which increases the average length of the singlets by ~ 1.5 compared with $\Delta_1/\epsilon = 1.8$ used in Secs. V and VI of the main text. Compared with the results from Fig. 3(a), Fig. 12(a) shows that this environment decreases the binding energy. In Fig. 12(b), we have further decreased it to $\Delta_1/\epsilon = 0.18$, and here we see that the red band completely disappears into the scattering continuum.

To further establish these results, we show in Fig. 13 the results from the same calculations as those in Fig. 10(c) but with $\Delta_1/\epsilon = 0.18$. While the results in Fig. 10 show indications of bound state formation, these do not, and we find a general trend that decreasing Δ_1/ϵ results in the iterative calculations converging for more crystal momenta in

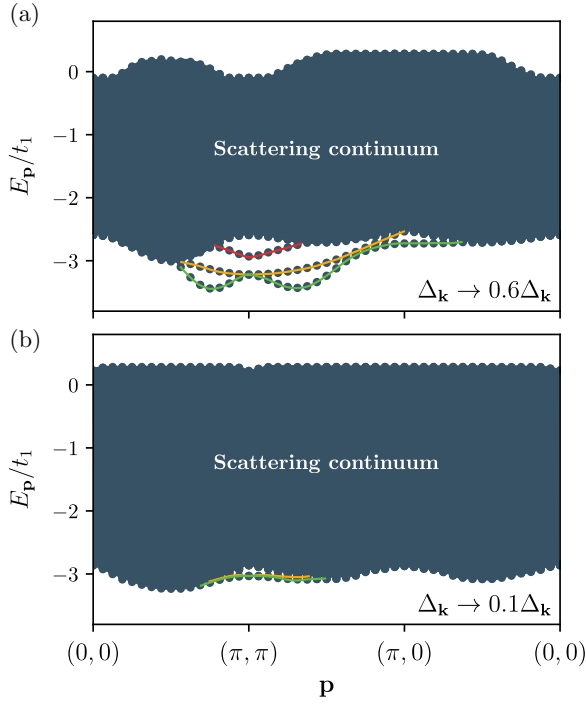


FIG. 12. With $J_1/t_1 = 0.6$ and Δ_1/Δ_2 constant, the energy spectrum for $\Delta_1/\epsilon = 1.08$ and $\Delta_1/\epsilon = 0.18$ is shown in (a),(b) respectively. The text in the panel states the overall scaling of the pairing potential. The calculations are performed for a system size of 32×32 . Red, yellow, and green lines highlight bound states below the scattering continuum.

the Brillouin zone. Altogether, this indicates that having a stiffer spinon spectrum with shorter singlets assists the formation of spinon-holon bound states, while systems with longer singlets promote separation of the spinon and the holon, yielding a broad spectral response.

APPENDIX D: GAUGE AVERAGING

In Secs. V and VI of the main text, we perform a gauge averaging to recover the full square lattice symmetry of

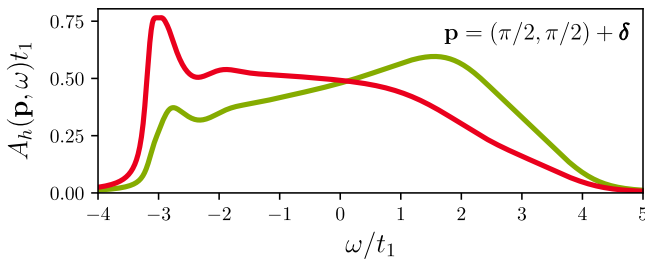


FIG. 13. In red, we plot the spectral function calculated with the iterative method described in Appendix B. We performed 50 iterations and found convergence. The co-propagating term is plotted in green, and the calculations are performed with $\Delta_1/t_1 = 0.18$, $J_1/t_1 = 0.6$, and $\delta = (2\pi/L, 2\pi/L)$, for a system size of 16×16 .

all observables. Here, we elaborate on this procedure and where the symmetry breaking originates.

As explained in the main text, the optimal mean-field description of the $\mathbb{Z}_2$ QSL is

$$\hat{H}_J^{\text{mf}} = \sum_{\mathbf{k}, \sigma} \epsilon_{\mathbf{k}} \hat{f}_{\mathbf{k}, \sigma}^\dagger \hat{f}_{\mathbf{k}, \sigma} + \sum_{\mathbf{k}} \left(\Delta_{\mathbf{k}} \hat{f}_{\mathbf{k}, \uparrow}^\dagger \hat{f}_{-\mathbf{k}, \downarrow}^\dagger + \text{h.c.} \right). \quad (\text{D1})$$

When projected onto the half-filled subspace, the Cooper pairs of the BCS wave function turn into the singlets covering the lattice. The presence of both $d_{x^2-y^2}$ and d_{xy} pairing apparently breaks reflection symmetry; for example, $x \rightarrow -x$ changes the relative sign of the two pairing components (as does $x \leftrightarrow y$). At half-filling, however, it is known that gauge-invariant observables are fully symmetric, as discussed in Refs. [37,61]. In particular, the relative sign of the two pairing terms is modified by the pure gauge transformation

$$\hat{f}_{i\alpha} \rightarrow \epsilon_{\alpha\beta} \hat{f}_{i\beta}^\dagger (-1)^{|i|}. \quad (\text{D2})$$

This is part of the $\text{SU}(2)$ gauge symmetry of the spin-liquid ansatz $\hat{\mathbf{S}}_i = \frac{1}{2} \hat{f}_i^\dagger \boldsymbol{\sigma} \hat{f}_i$, so at half-filling, for which all observables can be expressed in terms of spin operators, the parity noninvariance of $\hat{H}_J^{\text{mf}}$ is purely a gauge artifact.

However, in the larger Hilbert space of the t - J model, and with use of the $\text{U}(1)$ slave boson ansatz $\hat{c}_{i\alpha} = \hat{b}_i^\dagger \hat{f}_{i\alpha}$, the transformation in Eq. (D2) is no longer possible (because there is no corresponding transformation of the holon operators that leaves the electron invariant). Therefore, our theory describes a situation in which the parity symmetry becomes truly broken for physical observables.

Because the underlying t - J model has parity symmetry, the parity breaking must be spontaneous, and hence in repeated experiments the choice of the parity-broken state will vary from realization to realization. To account for this, we perform a gauge averaging over the different gauges of $\Delta_{\mathbf{k}}$. Specifically, we perform two calculations—one using $\Delta_{(k_x, k_y)}$ and the other using $\Delta_{(k_y, k_x)}$ —and then we take the average of the two results. For the relative wave function $\phi_{\mathbf{p}, \mathbf{r}}^n$ in Fig. 3(c), this means we compute $|\phi_{\mathbf{p}, \mathbf{r}}^n|^2$ for both gauges individually and then we take the average.

In principle, it is possible to choose a different formulation of the doped system which retains the full symmetry of the half-filled spin-liquid state, but this requires going beyond the $\text{U}(1)$ slave boson theory to the $\text{SU}(2)$ slave boson theory [91], in which there are two flavors of holon operators. We leave such an approach to future work.

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