

Confined and deconfined spinon excitations in the rectangular-lattice quantum antiferromagnetN. E. Shaik,¹ E. Fogh¹, B. Dalla Piazza,¹ B. Normand^{1,2}, D. A. Ivanov^{1,3} and H. M. Rønnow¹¹*Laboratory for Quantum Magnetism, Institute of Physics, [Ecole Polytechnique Fédérale de Lausanne \(EPFL\)](#), CH-1015 Lausanne, Switzerland*²*PSI Center for Scientific Computing, Theory and Data, CH-5232 Villigen-PSI, Switzerland*³*Institute for Theoretical Physics, [ETH Zürich](#), CH-8093 Zürich, Switzerland*

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Fractionalization remains one of the most fascinating manifestations of strong interactions in quantum many-body systems. In quantum magnetism, the existence of spinons—collective magnetic excitations that behave as quasiparticles with fractional quantum numbers—is proven in spin chains, but the criteria for their appearance in higher dimensions remain disputed. Motivated by experiments reporting the observation of spinons at high energies in the square-lattice Heisenberg antiferromagnet, we adopt the approach of extrapolating from where spinons are well defined. We study the dynamical properties of a Gutzwiller-projected wave function, the staggered-flux state, on a rectangular spin-1/2 Heisenberg lattice as a function of the spatial coupling ratio, $\gamma = J_y/J_x$. By studying the spectrum and the spinon separation distribution we show how, as the system evolves from one-dimensional (1D) towards 2D, the spinons become progressively more confined over most of reciprocal space, but remain deconfined at specific wave vectors.

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Introduction. The emergence of quasiparticles with fractional quantum numbers is a fascinating collective phenomenon. Notable examples include quarks confined as hadrons [1], fractionally charged quasiparticles in the quantum Hall effect [2], spin-charge separation of electrons in one-dimensional (1D) systems [3], solitons in polyacetylene-type molecules [4], and spinons in antiferromagnetic (AFM) spin-1/2 Heisenberg chains [5–7]. While the fractionalization of spin excitations in 1D, represented schematically in Figs. 1(a)–1(b), is clearly demonstrable in theoretical models, the existence of true spinons in higher dimensions remains a challenge. Ideal fractionalization, meaning on all energy scales, is present in certain 2D models that have yet to be realized faithfully in quantum magnetic materials, but in the spin-1/2 Heisenberg compounds known to date is replaced by conventional behavior as the two $S = 1/2$ fractions become confined into $S = 1$ magnons. While the search continues for materials displaying complete deconfinement as a consequence of sufficiently strong quantum fluctuations, for example when frustration effects act to suppress magnetic order as a competing ground state, a less strict criterion for fractionalization is the observation of spinonic fingerprints at finite energies.

The idea that magnetic correlations in 2D systems can be dominated by the quantum fluctuations clearly manifest in 1D was proposed originally in the context of high-temperature

superconductivity (HTS) [8]. The associated concepts, illustrated in Fig. 1(c), provided a starting point for the study of quantum spin liquids [9,10]. Although the ground state of the quantum ($S = 1/2$) square-lattice Heisenberg AFM relevant to HTS has long-ranged Néel order and sharp magnon excitations at low energies [11], which have been verified quantitatively in a number of materials [12–15], detailed spectroscopic measurements [13,16–22], still at base temperature, have found anomalous, continuum-like scattering at higher energies. Multiple theoretical studies have debated whether these results indicate extensive magnon bound-state formation [23–27] or a possible deconfinement of single magnons into fractional constituents [22,28–32].

In this Letter we perform a systematic analysis of fractionalization in the excitation spectrum of the $S = 1/2$ Heisenberg model on a rectangular lattice, i.e., with no frustration. To understand if and how the reports of deconfinement at $(\pi, 0)$ in the 2D limit are connected to complete deconfinement in 1D, we build on prior work establishing the validity of the staggered-flux state as an accurate description of the ground state of the rectangular-lattice model at all values of the spatial coupling ratio, $\gamma = J_y/J_x$. We analyze the excitation spectrum of the staggered-flux state by using a Monte Carlo method to compute the transverse dynamical structure factor and spinon pair separation. Starting from a quantitatively accurate description of the 1D limit, we demonstrate that increasing γ causes a systematic spreading of confinement in reciprocal space, but explicitly not in energy, until in the square-lattice limit ($\gamma = 1$) the deconfined nature remains only at the $(\pi, 0)$ points.

We first review the concept of spin fractionalization with the schematic images of Fig. 1. When a single spin is flipped in a 1D Néel state, it can be regarded as two domain walls whose separation costs no further energy [Fig. 1(a)]. If the

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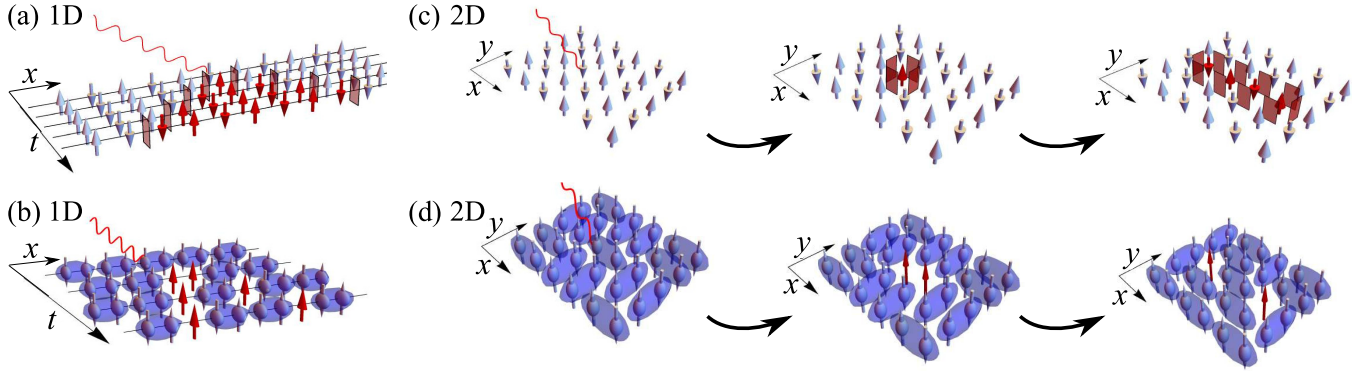


FIG. 1. Schematic representations of the propagation of spin-flip excitations in low-dimensional quantum spin systems. (a) A spin flip is created by scattering an incident neutron (wavy red line). In a 1D Néel state it creates two domain walls, represented by the semitransparent red rectangles. Quantum spin fluctuations permit the propagation of domain walls at subsequent time steps (t), whence each can be interpreted as a spinon. (b) A spin flip in the nearest-neighbor valence-bond state (dimer singlets are represented by purple ellipses) on a chain creates one dimer triplet (represented by parallel spins with no ellipse). These two uncorrelated spins may propagate at no additional energy cost on the singlet background. (c,d) Analogous representations on the AFM square lattice, where now x and y are two spatial dimensions and time steps are represented by the sequence of panels. The propagation of two domain walls in a Néel state would create two lines of ferromagnetic bonds, leading instead to confinement (c). By contrast, in a valence-bond state (d) the two freed spins can still propagate at no extra cost.

macroscopic singlet ground state of a Heisenberg spin chain is visualized as a superposition of short-ranged dimer singlets [only nearest-neighbor valence bonds are shown in Fig. 1(b)], a single spin excitation creates a dimer triplet whose two constituent spins are again free to delocalize. These $S = 1/2$ excitations were established theoretically by the Bethe Ansatz [33] and have been observed in experiment [5–7]. However, extending the domain-wall image to a 2D Néel state [Fig. 1(c)] shows clearly why a spin flip should preserve $S = 1$ (magnonic) character, with the 1D string of FM bonds creating linear confinement of putative spinons. This has led to the notion that geometrical frustration strong enough to create a disordered (quantum spin-liquid [9]) ground state [Fig. 1(d)] is required to permit fractionalization in 2D.

From all the reports of scattering anomalies at the boundaries of the magnetic Brillouin zone in $S = 1/2$ square-lattice materials [13,16–22], the system best realizing the minimal, nearest-neighbor-only AFM Heisenberg model is copper formate tetradeuterate (CFTD) [13]. Detailed measurements have revealed [20,22] that the magnon branch of a renormalized spin-wave theory survives as a sharp mode everywhere within and on the boundary of the Brillouin zone, except at the wave vectors $(\pi, 0)$ and $(0, \pi)$, where it loses spectral weight in favor of an apparent scattering continuum at higher energies. The most detailed theoretical and numerical studies of the spectrum, whether based on magnon bound states [26,27] or on fractionalization [22,31,32], have also highlighted the special nature of the $(\pi, 0)$ and symmetry-related points.

Staggered-flux state on a rectangular lattice. It was shown in Ref. [22] that the staggered-flux state [29,30] on the square lattice exhibits continuum-like scattering and excitations akin to deconfined spinons at the $(\pi, 0)$ points [34]. To establish whether a spinon interpretation is viable, and if so to connect this putative finite-energy deconfinement in 2D with the complete fractionalization observed in 1D, we therefore adopt the staggered-flux Ansatz. We use it to perform an explicit study of the spectrum of the Heisenberg model on the rectangular

lattice (represented in the inset of Fig. 4),

$$\mathcal{H} = \sum_{\mathbf{i}} J_x \mathbf{S}_{\mathbf{i}} \cdot \mathbf{S}_{\mathbf{i}+\hat{x}} + J_y \mathbf{S}_{\mathbf{i}} \cdot \mathbf{S}_{\mathbf{i}+\hat{y}}, \quad (1)$$

by altering the coupling ratio, $\gamma = J_y/J_x$, to traverse smoothly from a quasi-1D to a 2D system [35–38].

The staggered-flux state is obtained from the mean-field decoupling of an Ansatz describing the system in terms of fermionic spinons, as we summarize in Sec. S1A of the Supplemental Material (SM) [39]. The mean-field state, $|\psi_{\text{SF}}\rangle$, allows double site occupancy and a Gutzwiller projection [40], $|\psi_{\text{GS}}\rangle = P_G |\psi_{\text{SF}}\rangle$, is performed to eliminate doubly-occupied sites and thus obtain the ground state in the physical state space as a variational wave function. In Ref. [41], we optimized the staggered flux in order to discuss the ground state of this rectangular-lattice Heisenberg model [Eq. (1)] as a function of γ . Although the staggered-flux state cannot reproduce the exact ground state of the square-lattice model, most notably lacking an ordered moment, we will show that it offers a valuable starting point for constructing this state in the sense that its finite-energy features, including fractional excitations, provide an excellent account of the spectral function.

In the variational approach one approximates the physical excitations by projected fermionic excitations [29,30]. Following Ref. [22], we construct transverse (spin-flip) excitations as linear combinations of fermionic particle-hole pairs,

$$|\psi_{\mathbf{q}}^n\rangle = \sum_{\mathbf{k}} \phi_{\mathbf{k}\mathbf{q}}^n |\mathbf{k}, \mathbf{q}\rangle, \quad \text{with } |\mathbf{k}, \mathbf{q}\rangle = P_G d_{\mathbf{k}\uparrow}^{+\dagger} d_{\mathbf{k}-\mathbf{q}\downarrow}^- |\psi_{\text{SF}}\rangle, \quad (2)$$

where $d_{\mathbf{k}-\mathbf{q}\downarrow}^-$ destroys a down spin in the lower band, $d_{\mathbf{k}\uparrow}^{+\dagger}$ creates an up spin in the upper band, and the coefficients $\phi_{\mathbf{k}\mathbf{q}}^n$ are obtained by diagonalizing the Hamiltonian (1) projected onto the nonorthogonal states $|\mathbf{k}, \mathbf{q}\rangle$; further details are provided in Sec. S1B of the SM [39]. We adopt a Monte Carlo sampling technique to solve the associated generalized eigenvalue problem (Sec. S1C) and hence to compute the transverse

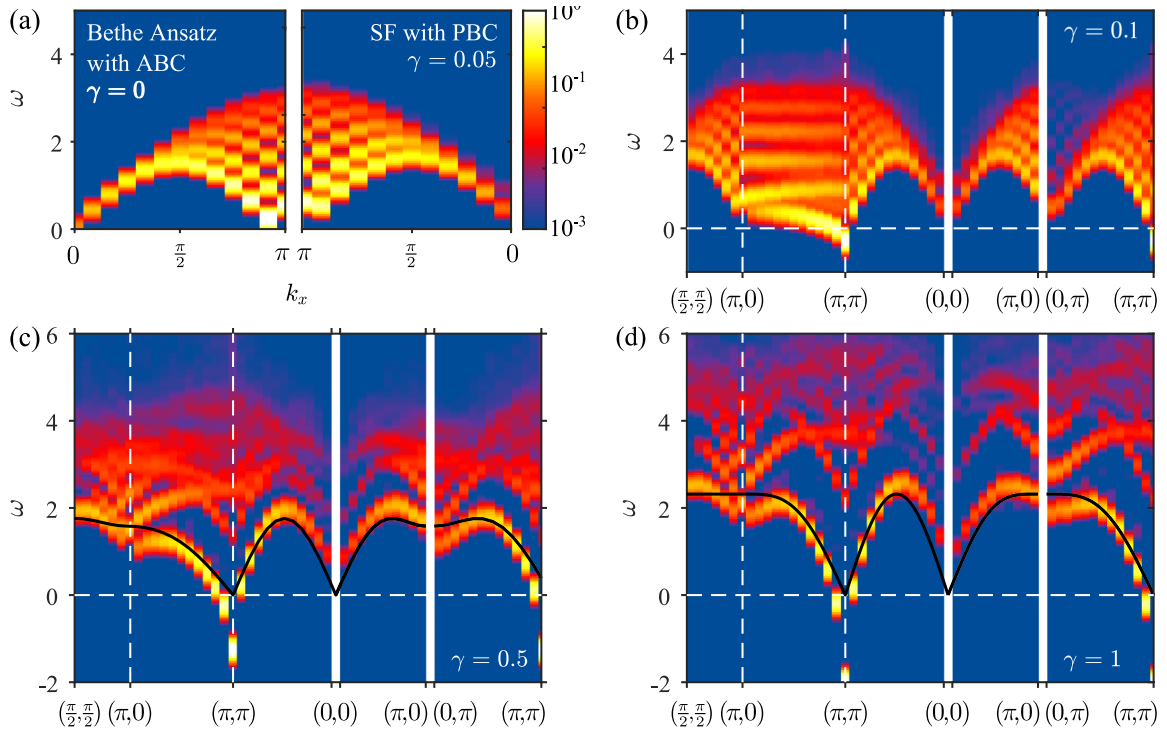


FIG. 2. Spectral functions on the rectangular lattice. (a) Comparison between the transverse dynamical structure factors computed for a chain of $L = 24$ sites (i.e., $\gamma = 0$) using the algebraic Bethe Ansatz (left) and the staggered-flux (SF) state with $\gamma = 0.05$ (right). ω is expressed in units of J_x . (b)–(d) Transverse dynamical structure factors calculated with PBCs in x and ABCs in y , on a selected $\mathbf{q}$ -space path (inset, Fig. 4) for coupling ratios $\gamma = 0.1$ (b), $\gamma = 0.5$ (c), and $\gamma = 1$ (d) at system size $L = 24$. Black lines show the dispersion obtained from a self-consistent spin-wave theory (Sec. S3 of the SM [39]). The asymmetry between $(\pi, 0)$ and $(0, \pi)$ in panel (d) is due to the BCs.

dynamical structure factor (TDSF), which we approximate as

$$S^\pm(\mathbf{q}, \omega) = \sum_n |\langle \psi_{\mathbf{q}}^n | S_{\mathbf{q}}^\pm | \psi_{\text{GS}} \rangle|^2 \delta(\omega - E_{\mathbf{q}}^n + E_{\text{GS}}), \quad (3)$$

where E_{GS} and $E_{\mathbf{q}}^n$ are respectively the variational energies of the ground and excited states. Thus, we perform Monte Carlo simulations on systems of size $N = L \times L$, with L up to 28, working in the space of all singly occupied states with $N/2 + 1$ up and $N/2 - 1$ down spins, and using a statistical average of the energies of the diagonalized Hamiltonian to obtain a suitably $\mathbf{q}$ -resolved spectrum. The staggered-flux wave function is ill-defined at its nodal points, $\mathbf{k} = (\pm\pi/2, \pm\pi/2)$, where numerator and denominator terms vanish, and we choose to avoid these by keeping antiperiodic boundary conditions (ABCs) in y while working with both ABCs and periodic BCs (PBCs) in x . This approach is valuable both for comparison with the exact solution in 1D and for studying finite-size effects in our calculations (Sec. S2 of the SM [39]).

To characterize the spatial profile of these excitations at each $\mathbf{q}$, we introduce the weighted spatial spinon density,

$$\rho^q(\mathbf{r}) = \sum_n \frac{|\langle \psi_{\mathbf{q}}^+(\mathbf{r}) | \psi_{\mathbf{q}}^n \rangle \langle \psi_{\mathbf{q}}^n | \psi_{\mathbf{q}}(0) \rangle|^2}{\langle \psi_{\mathbf{q}}^+(\mathbf{r}) | \psi_{\mathbf{q}}^+(\mathbf{r}) \rangle}, \quad (4)$$

where $|\psi_{\mathbf{q}}^+(\mathbf{r})\rangle$ represents a pair of spinons induced at momentum transfer $\mathbf{q}$ with a separation $\mathbf{r} = (x, y)$ (Sec. S1B of the SM [39]). As a means of distinguishing a possible spinon delocalization in 2D from the 1D delocalization we approach

in the small- γ limit, we define the quantities

$$D_x(\mathbf{q}) = \sqrt{\sum_{\mathbf{r}} x^2 \rho^q(\mathbf{r})} \quad \text{and} \quad D_y(\mathbf{q}) = \sqrt{\sum_{\mathbf{r}} y^2 \rho^q(\mathbf{r})}. \quad (5)$$

These measure the RMS value of the spinon separation in one lattice direction and hence serve to gauge the degree and nature of spinon deconfinement.

Results. Diagonalizing the projection of $\mathcal{H}$ onto the $|\mathbf{k}, \mathbf{q}\rangle$ basis yields N discrete energies at each value of $\mathbf{q}$. We set $J_x = 1$ as the unit of energy and convolve the discrete spectra with Gaussians of width $\sigma = 0.1J_x$ to obtain the TDSFs we show in Fig. 2. This resolution is consistent with our system sizes (Sec. S2 of the SM [39]) and, because our focus is away from the lowest-energy regions, does not generate spurious results for small γ values.

The prerequisite for our extrapolation across the rectangular lattice is to benchmark the validity of the staggered-flux state as a description of the spectrum in the 1D limit, where ideal (fully deconfined) spinons form the basis for the exact solution [33]. Figure 2(a) compares the TDSF calculated using the algebraic Bethe Ansatz [42] with that of a staggered-flux state at $\gamma = 0.05$ (the lowest coupling we use) and $q_y = 0$. We find that the agreement is not only excellent, but quantitatively accurate in energy and intensity when an L -site chain is compared with an $L \times L$ rectangular lattice. The $\gamma = 0.05$ spectrum lies higher in energy by precisely the finite interchain coupling. The fermionic nature of the staggered-flux wave function makes the Bethe Ansatz solution

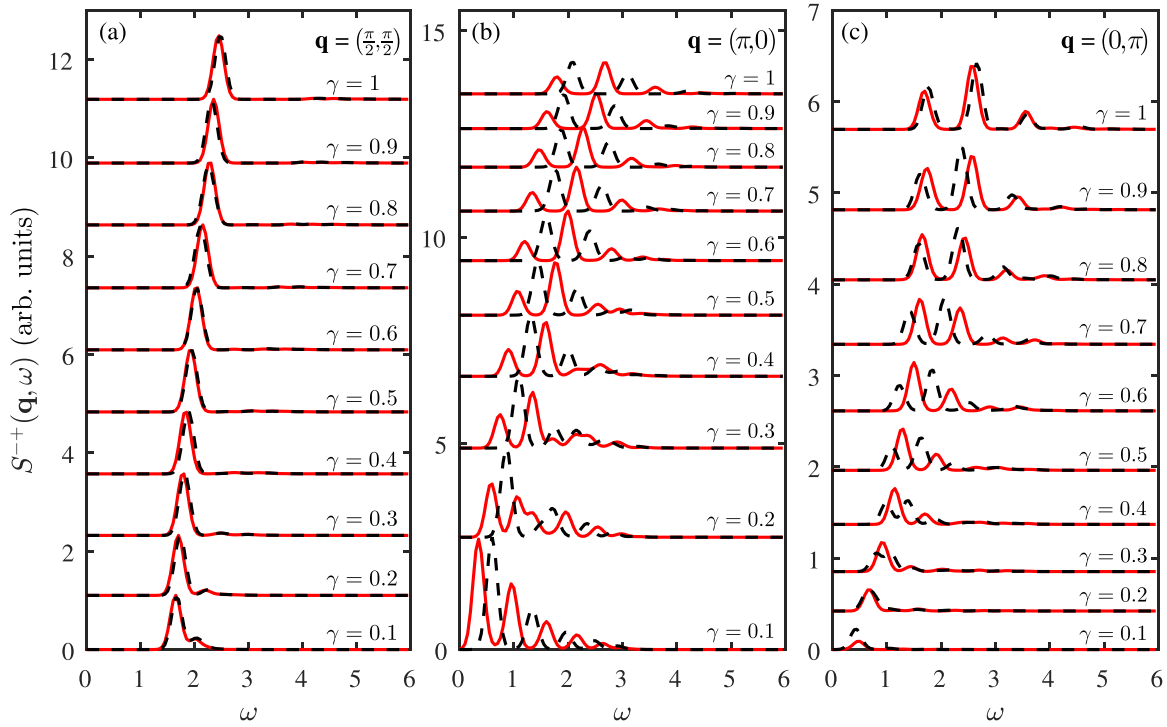


FIG. 3. Evolution of the TDSF with γ . TDSF calculated with $L = 28$ at $(\pi/2, \pi/2)$ (a), $(\pi, 0)$ (b), and $(0, \pi)$ (c). The black lines represent calculations performed with PBCs in x and ABCs in y , the red lines ABCs in both directions.

with ABCs equivalent to the staggered-flux state with PBCs and vice versa [41], which is reflected in the $2\pi/L$ shift in the position of the Goldstone mode near $k_x = \pi$ in Fig. 2(a). This confirmation lays the foundation for a systematic study of spinon confinement effects when γ is used to bring the system from 1D to 2D.

Figures 2(b)–2(d) show the TDSF for the three coupling ratios $\gamma = 0.1, 0.5$, and 1 to capture the evolution of the spin dynamics with increasing two-dimensionality. The unphysical negative energies at (π, π) reflect how the staggered-flux state, with no long-ranged order, fails to capture the Goldstone mode and the neighboring low-energy spectrum. At all other $\mathbf{q}$ -points we observe how a quasicontinuum at small coupling ratios [Fig. 2(b)] evolves into sets of discrete modes [Fig. 2(d)]. The loss of spectral weight in the high-energy modes with increasing γ occurs everywhere except at $(\pi, 0)$, where these modes continue to provide almost half of the spectral weight at $\gamma = 1$. In Sec. S2 of the SM [39] we present a finite-size comparison of different $\mathbf{q}$ -points in this case. In Figs. 2(c) and 2(d) we show also the magnon branch obtained from a self-consistent spin-wave theory [43], which provides an energetic renormalization factor, $Z_c(\mathbf{q})$, to linear spin-wave theory as described in Sec. S3 of the SM [39]; away from the negative-energy points we find a rather good match to the low-energy mode of the staggered-flux state.

Figure 3 shows the evolution of the TDSF calculated at $L = 28$ for the three high-energy points $(\pi/2, \pi/2)$, $(\pi, 0)$, and $(0, \pi)$. At $(\pi/2, \pi/2)$ we observe the generic behavior of weak additional modes above the lowest one at low γ , which lose all their weight as the spectrum evolves towards 2D to contain only a single zone-boundary magnon [Fig. 3(a)]; the insensitivity of the peak energies to the BCs is also one

indicator of robust confinement. At $(\pi, 0)$, by contrast, the additional peaks retain significant weight even at $\gamma = 1$, and the choice of BCs shifts their positions by an amount similar to half their separation [Fig. 3(b)], reflecting the deconfinement we characterize below. Similar behavior is observed at $(0, \pi)$ [Fig. 3(c)], although here the spectral weight grows from zero at the 1D limit to fewer, stronger “continuum” modes due to our system size. Extending the insight of Fig. 3 back to Figs. 2(b)–2(d), our results suggest that in the thermodynamic limit the spectrum forms a single, sharp magnon-like mode as $\gamma \rightarrow 1$, at all $\mathbf{q}$ -points other than $(\pi, 0)$ and $(0, \pi)$; there we find that a series of discrete modes persists, whose number increases with L (Sec. S2 of the SM [39]), suggesting the formation of a scattering continuum.

To interpret the line shapes in the TDSF, in Fig. 4 we show the two RMS spinon separations $D_x(\mathbf{q})$ and $D_y(\mathbf{q})$ over the high-symmetry directions of $\mathbf{q}$ -space for the full range of coupling ratios. As γ is increased, $D_x(\mathbf{q})$ decreases at most $\mathbf{q}$ -points, reflecting a progressive spinon confinement in the chain direction as a consequence of interchain coupling [Fig. 4(a)]. By contrast, $D_x(\mathbf{q})$ provides an excellent illustration of how γ has no effect on confining spinon pairs at wave vectors $(\pi, 0)$ and $(0, \pi)$. Concerning the analogous result at (π, π) and $(0, 0)$, these low energies are where the staggered-flux state without in-built AF order [22] captures neither the Goldstone mode nor, presumably, the confining effect of the ordered state on the spinons, and hence we do not consider these points further. For further insight into the meaning of deconfinement at $(0, \pi)$ in this geometry, Fig. 4(b) shows that $D_y(\mathbf{q})$ is almost invariant with γ , indicating how the trivial spinon localization caused by small interchain coupling turns into a real localization due to the 2D character of the system,

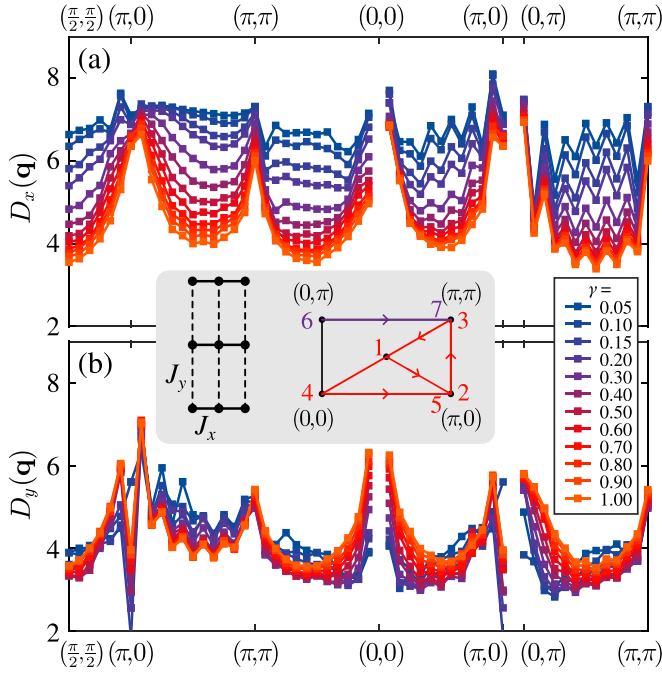


FIG. 4. RMS spinon pair separation. Pair-separation parameters $D_x(\mathbf{q})$ (a) and $D_y(\mathbf{q})$ (b) calculated on a system of $L = 24$ with PBCs in x , shown on the same $\mathbf{q}$ -space path as in Fig. 2 for different coupling ratios, γ . The inset shows a schematic representation of the rectangular lattice with superexchange couplings J_x and J_y , and of the corresponding reciprocal space; numbers 1–7 indicate the order in which the $\mathbf{q}$ -space path is followed.

with the exception of the growth in RMS separation around the $(0, \pi)$ and $(\pi, 0)$ [and (π, π) and $(0, 0)$] points.

For a clear visualization of differing behavior in the spinon pair separation, we focus on the points $(\pi/2, \pi/2)$ and $(\pi, 0)$ to show the RMS separations as functions of γ in Fig. 5. In the subsidiary panels we show the corresponding spinon pair-density distributions at $\gamma = 0.1, 0.5$, and 1 on a 28×28 lattice. At $(\pi/2, \pi/2)$, we observe a gradual decrease of $D_x(q)$ and a clear localization of pair density around $r = 0$ that has almost saturated at $\gamma = 0.5$. As for all $\mathbf{q}$ -points other than the exceptions visible in Fig. 4, this is the characteristic sign of spinon confinement, even though the ground state (the staggered-flux state) has no magnetic order. Again the $(\pi, 0)$ point is completely different, with $D_x(\mathbf{q})$ remaining flat and the system-scale 1D distribution turning into a system-scale 2D distribution, indicating that the spinons remain fully deconfined.

Conclusions. We have studied the crossover from 1D to 2D physics in the $S = 1/2$ Heisenberg model with only nearest-neighbor coupling and no frustration. We base our investigation on a Gutzwiller-projected variational wave function that yields a staggered-flux ground state, and thus we obtain results that are highly indicative but not definitive. By comparison with the Bethe Ansatz solution, we demonstrate that the staggered-flux state provides a quantitatively accurate description of the excitation spectrum in the 1D limit, reproducing the des Cloizeaux-Pearson-Faddeev spinon continuum [44,45]. Increasing the interchain coupling to the 2D limit causes a systematic vanishing of the spectral weight at all higher en-

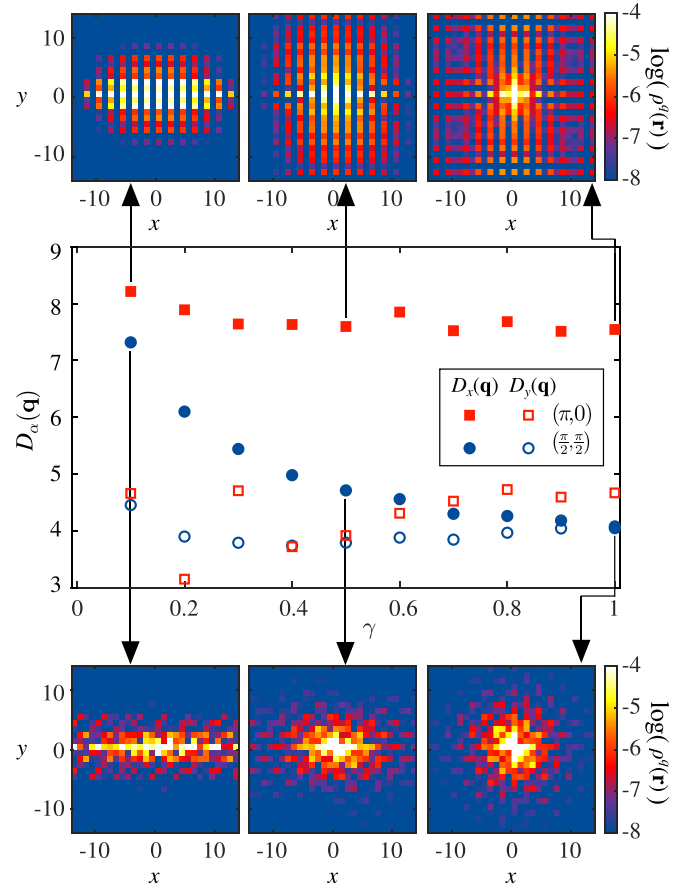


FIG. 5. Spinon pair separation on the rectangular lattice. $D_x(\mathbf{q})$ and $D_y(\mathbf{q})$ calculated on a system of $L = 28$ with PBCs in x , shown as functions of γ at $\mathbf{q} = (\pi, 0)$ and $\mathbf{q} = (\pi/2, \pi/2)$. The subpanels show the corresponding spinon pair-separation distributions, $\rho^{\mathbf{q}}(\mathbf{r})$.

ergies at almost all wave vectors, leaving only a well-defined magnon mode that can be considered as a pair of confined spinons. Only the reciprocal-space points $(\pi, 0)$ and $(0, \pi)$ appear resistant to confinement, with their spectrum retaining a continuum form above its lower edge and their spinon-pair excitations remaining distributed across the entire system.

Our results reinforce the deduction [22] that deconfinement in the rectangular lattice is a reciprocal-space phenomenon, and is not connected directly to the magnon energy. For more insight into the role of kinematic factors in 2D systems, as opposed to the degree of quantum fluctuations alone, one qualitative comparison for the square lattice is the honeycomb lattice, which is also unfrustrated (bipartite) such that a $S = 1/2$ Heisenberg model has long-range order in the ground state. Experimental [46] and numerical [47,48] studies indicate that deconfinement is more widespread around the zone boundary on the honeycomb lattice, suggesting less sensitivity to purely kinematic factors than on the square lattice, where only two deconfined points survive. A more quantitative study of this question could perhaps be performed with J_1 - J_2 square lattice [49,50], where the geometry is not a variable.

The confinement process we compute appears as a smooth crossover, with a gradual suppression of spinonic behavior as a function of γ at all “conventional” wave vectors (Figs. 4 and

5), as opposed to an abrupt one. Because our studies consider finite systems, they do not exclude that spinon confinement occurs in the thermodynamic limit at all finite γ and all $\mathbf{k}$, other than $(\pi, 0)$ and $(0, \pi)$, but they do show that there is no evidence for a confinement transition at some finite γ .

As already noted, the (π, π) point is a weakness of the staggered-flux wave function, with the poor treatment of the low-energy dispersion (Fig. 2) being a consequence of the missing ordered moment. However, enforcing long-ranged order in the staggered-flux framework results in spinon confinement at all energies [22] and hence a poor description of the measured spectrum. In finding a balance between these semiclassical and quantum properties, studies of variational states in 2D found long-range order to be a rather minor modification to a state of otherwise robust quantum fluctuations [51]. For spectroscopic studies probing finite energies and finite temperatures giving finite excitation linewidths, this implies that the part of the wave function ensuring the

low-energy, long-wavelength limit may be of little consequence for the spectrum over most of the Brillouin zone. Here we have quantified the spinonic character of the rectangular-lattice spectrum, demonstrating the relevance of spinons at energies above J_x , but nevertheless with finite confinement lengths, which decrease with increasing γ , at all wave vectors other than the fully deconfined points $(\pi, 0)$ and $(0, \pi)$.

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Data availability. The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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