

Cooper-pair localization in the magnetic dynamics of a cuprate ladder

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We investigate the spin dynamics of the cuprate ladder $\text{Sr}_{2.5}\text{Ca}_{11.5}\text{Cu}_{24}\text{O}_{41}$ to elucidate the behavior of its intrinsically doped holes. By combining high-resolution inelastic neutron spectroscopy with density-matrix renormalization group calculations, we obtain a detailed and quantitative description of its collective magnetic excitations within a single-band framework. Remarkably, we observe an absence of magnetic signatures associated with unpaired charges, suggesting that the doped holes form strongly bound, localized Cooper pairs. Indeed, while the standard single-band Hubbard model fails to reproduce the observed excitation spectrum, a minimal extension incorporating a strong attractive nearest-neighbor interaction quantitatively accounts for the experimental results. Our findings underscore the necessity of including interactions beyond conventional magnetic mechanisms in driving charge pairing when the system is modeled using the single-band Hubbard model. Given the close correspondence between ladder systems and two-dimensional cuprates, these results may have important implications for pairing phenomena in square-lattice cuprate materials.

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I. INTRODUCTION

Cuprate ladders have been studied extensively since the 1990s as stripped-down models of high-temperature (high- T_c) superconductivity [1,2]. These materials share many similarities with the two-dimensional cuprates, including superconductivity under pressure, yet are more amenable to theoretical modeling. Ladder behavior has also been invoked as a key component of recent theories of hole-doped planar cuprates based on intertwined stripes and pair-density-waves [3,4]. However, the details of the underlying pairing mechanism in cuprate ladders remain an open question. While the general belief is that strong electronic correlations play a crucial role, research on doped cuprate ladders has generated perplexing and contradictory outcomes when comparing theory and experiments, to the extent that even the

precise doping levels of these materials have been a subject of controversy [2].

An essential challenge in understanding correlated electron materials is to identify minimal Hubbard Hamiltonians that capture the high energy excitations associated with the underlying bare interactions, as well as the low-energy collective modes. The measured structure of the magnetic excitations thus imposes a stringent test for candidate models. Motivated by this, here we reexamine the case of $\text{Sr}_{2.5}\text{Ca}_{11.5}\text{Cu}_{24}\text{O}_{41}$ (SCCO). This material has long been the focal point of research as its magnetism and strong correlations are believed to be responsible for the formation of a possible charge-density-wave (CDW) [5] and pressure-induced superconductivity [6]. Here, we undertake high-resolution inelastic neutron scattering (INS) measurements with matching density-matrix renormalization group (DMRG) [7] calculations of the dynamical spin structure factor of an extended single-band Hubbard model. Through this, we pinpoint the principal interactions of a low-energy effective model for this prototypical ladder system and shed light on the behavior of doped holes, within the framework of an effective single-band description.

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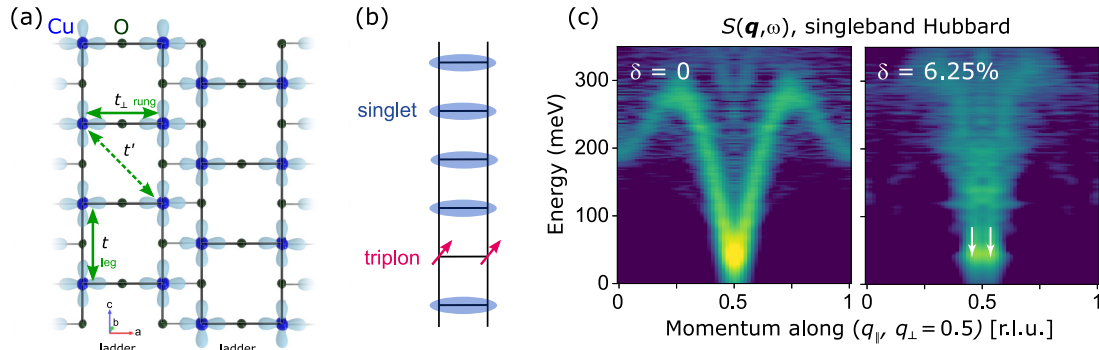


FIG. 1. Ladder structures and magnetic dynamics of the cuprate ladder $\text{Sr}_{2.5}\text{Ca}_{11.5}\text{Cu}_{24}\text{O}_{41}$. (a) A sketch of the ladder structure showing both the copper $3d_{x^2-y^2}$ and oxygen $2p_\sigma$ orbitals. The strong hybridization between the Cu and O orbitals leads to a strong superexchange coupling along both the ladder legs and rungs, while the offset geometry of the individual ladders results in a weak interladder coupling. In a down-folded single-band Hubbard ladder description, these orbitals lead to hopping along the leg (t), rung ($t_\perp$), and diagonal direction (t'), as indicated. (b) The expected one-triplon magnetic excitations for an undoped ladder with spin singlets on all rungs. (c) The expected magnetic excitation spectrum $S(\mathbf{q}, \omega)$ for a half-filled (undoped, $\delta = 0$) and $\delta = 6.25\%$ hole-doped Hubbard ladder with on-site interactions only, obtained from DMRG. The simulations show that doping the model produces incommensurate low-energy excitations, as indicated by the white arrows. As we will show, the experimental data for $\text{Sr}_{2.5}\text{Ca}_{11.5}\text{Cu}_{24}\text{O}_{41}$ more closely resembles the undoped case, despite the fact that the ladder subsystem is doped.

The SCCO family comprises Cu_2O_3 ladder layers separated by layers of Sr/Ca ions and CuO_2 chains, which serve as charge reservoirs. The Cu_2O_3 planes form a trellis lattice of spin-1/2 Cu sites, as shown in Fig. 1(a) [8]. Here, a subset of Cu ions are connected by 180° bonds to form two-leg ladders, with strong superexchange interactions mediated by O ions. Neighboring ladders in the plane are displaced by $c/2$, where c is the Cu–Cu distance along the leg, and connected by 90° bonds that are geometrically frustrated. This structure leads to a weak effective interladder coupling such that the material’s low-energy sector is dominated by contributions from largely independent ladder subsystems [9,10].

In the absence of doping (e.g., in $\text{La}_4\text{Sr}_{10}\text{Cu}_{24}\text{O}_{41}$), cuprate ladders are charge-transfer insulators hosting Cu $3d^9$ ions in a quantum cooperative-paramagnetic state [9]. Their ground state consists of a resonant superposition of singlet spin-pairs with short-range entanglement and gapped triplon modes [see Fig. 1(b)]. This physics is most easily understood by examining the strong-rung-coupling limit, where the dominant Heisenberg exchange coupling $J_\perp$ leads to a ground state with strong spin singlet character on each rung [1,11]. A finite energy Δ_T is required to break such a singlet and form a triplet (T) excited state carrying spin $S = 1$ and charge 0 (i.e., a triplon quasiparticle). For cuprate ladders like SCCO, the leg and rung couplings are comparable, $J \approx J_\perp$, and the ground state includes pairs of singlets that resonate between different configurations to decrease energy. Nevertheless, the excitation spectra of these materials can be well understood in terms of the same triplon quasiparticles inferred in the limit of strong-rung-coupling limit [11,12].

All excitations can be classified by parity with respect to reflections about the centerline of the ladder: states with an odd (even) number of triplons have odd (even) parity. Consequently, the INS intensity of one-triplon excitations, associated with the $q_\perp = \pi$ wavevector in units of the inverse lattice constant $1/c$, wavevector, is modulated as $0.5[1 - \cos(q_\perp)]$, while the two-triplon scattering intensity, associated with the $q_\perp = 0$ wavevector, is modulated as

$0.5[1 + \cos(q_\perp)]$ [9]. (From this point forward, all momenta will be given in units of the inverse lattice constant or reciprocal lattice units.) INS measurements on the undoped ladder material $\text{La}_4\text{Sr}_{10}\text{Cu}_{24}\text{O}_{41}$ confirm the expected singlet ground state, gapped triplon excitations, and parity behavior [9]. The triplon dispersion is characterized by a highly dispersive mode with an energy gap $\Delta_T \approx 30$ meV, which is lower than the spin gap expected for a nearest-neighbor Heisenberg Hamiltonian. The reduction of the spin gap can be captured by including four-spin interactions, such as the cyclic exchange J_{cyc} , as well as to further-neighbor exchange processes, which arise at order t^4/U^3 in a strong-coupling expansion of the Hubbard model (with U the on-site repulsion and t the hopping amplitude). The necessity of including such terms provides indirect evidence for substantial charge fluctuations, as the ratio $U/t \simeq 8$ that best reproduces the triplon dispersion is relatively small [9].

The SCCO family is intrinsically hole-doped with a formal valence on the copper sites of $+2.25$. While the majority of the 0.25 holes per Cu site reside in the chains, some holes are present in the ladder subsystem even without Ca doping [2,13]. Although substitution of Ca for Sr does not alter the formal valence, the associated chemical pressure transfers additional holes to the ladders from the chains; however, the precise amount of charge transfer is still being debated [2,14]. Nevertheless, different experimental approaches consistently place the ladder doping in the range $\delta = 0.06 - 0.21$ for the heavily Ca-doped systems. Moreover, Raman scattering [15,16], electrical conductivity [15,17], and x-ray diffraction measurements [5,18] have all conclusively shown ladder charge order across Ca concentrations with superconductivity ($x = 11-13$), consistent with the presence of doped holes in the ladder subsystem.

Hubbard ladder calculations show that doped holes form pairs [8,11,19,20], which either exhibit charge ordering or enter a superconducting state, depending on the interaction details. Doped holes will also affect the low-energy magnetic response, shown in Fig. 1(c) for the case of the single-band

TABLE I. Chopper frequencies for the various SEQUOIA settings, run in high-flux mode.

E_i (meV)	$T0$ (Hz)	Fermi 1 (Hz)
30	60	120
60	90	180
120	90	240
300	120	420
450	150	480

Hubbard model (see Sec. III for further details). In this case, the changes in the magnetic response can be easily understood by recognizing that each doped hole generates an antiphase boundary in the antiferromagnetic correlations [21]. This leads to incommensurate magnetic fluctuations at $q_{\parallel} = \pi(1 \pm \delta)$ ($q_{\perp} = \pi$), where δ is the effective hole doping of the ladder, as indicated by the white arrows in Fig. 1(c) [22]. However, as demonstrated in this study, we observe a remarkable absence of these incommensurate fluctuations in the INS data, despite the presence of doped holes in the ladder subsystem. We will show that this observation implies the presence of strongly localized Cooper pairs that preserve the dominant commensurate fluctuations at $q_{\parallel} = \pi$ via annihilation of opposite antiphase boundaries. The observation of pairing stronger than that generated by the bare repulsive Hubbard interaction suggests that additional mechanisms—such as ionic displacements—play a decisive role in stabilizing high- T_c

superconductivity, assuming that the single-band Hubbard model provides an adequate description of these materials.

II. EXPERIMENTAL RESULTS

A. Sample syntheses

Single crystals of $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$ ($x = 11.5$) (see also Fig. 1 within the Supplemental Material [24]) were grown in an image furnace using the traveling-solvent method following the procedure described in Ref. [25]. As starting materials, we used CaCO_3 and SrCO_3 (purity better than 3N5) from Alpha Aesar and CuO (purity 4N) from Sigma-Aldrich. Parts of all grown crystals were checked by x-ray powder diffraction and verified to have a single phase. The single crystallinity of the sample was checked by the E5 four-circle neutron diffractometer (Helmholtz Zentrum Berlin, Germany).

B. Inelastic neutron scattering measurements

We measured the neutron scattering spectrum of SCCO using the SEQUOIA spectrometer [26,27] at the ORNL SNS [28]. We mounted a 5-gm single crystal in the $0KL$ plane with the ladder legs (c direction) extending in the horizontal plane and the ladder rungs (a direction) extending in the vertical plane (Fig. 1). We measured the spectra with incident energies of 30, 60, 120, 300, and 450 meV in a fixed orientation with the ladder legs perpendicular to the incident beam. The instrument settings for each energy configuration are given in Table I. Measurements were carried out at 5 and 120 K (below

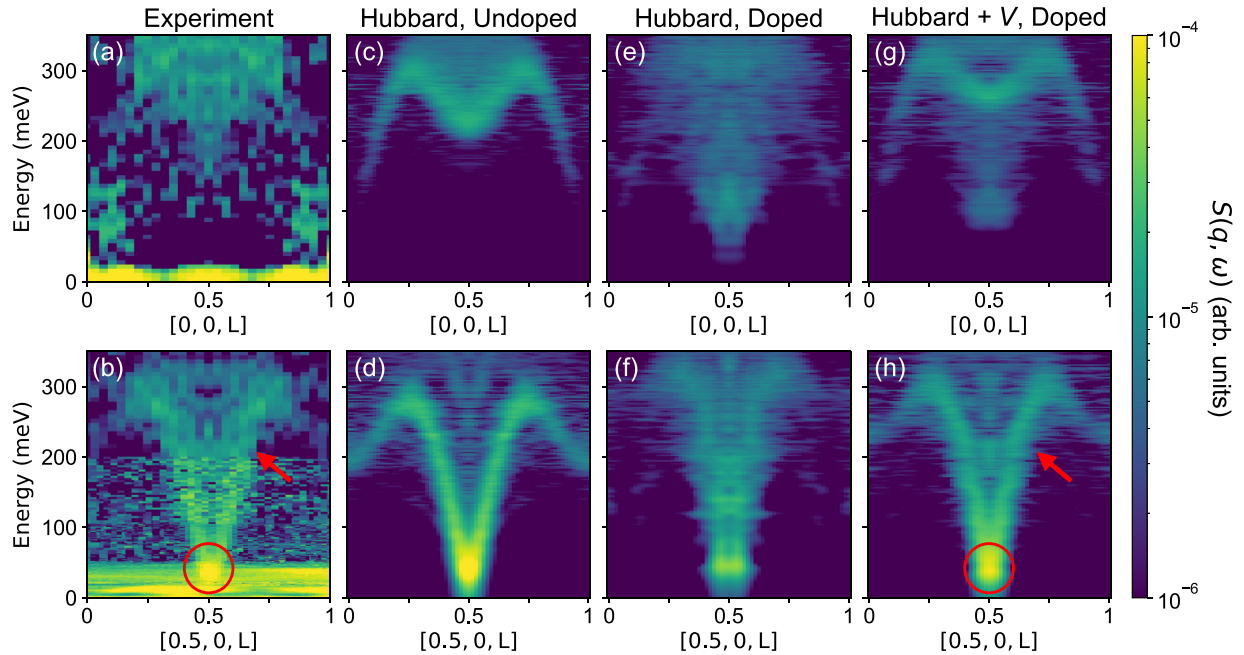


FIG. 2. A comparison of the measured spin-excitation spectrum and predictions from an extended Hubbard model. (a)(b) The measured INS spectra along the $\mathbf{q} = (q_{\perp}, 0, L)$ direction for $q_{\perp} = 0$ and 0.5 r.l.u., respectively, where the lattice spacing has been taken to be the Cu-Cu distance along the ladder leg. (c)(d) The dynamical spin structure factor $S(\mathbf{q}, \omega)$ obtained from DMRG calculations for an undoped Hubbard Hamiltonian without any nearest-neighbor interactions ($V = 0$). (e)(f) DMRG results for $S(\mathbf{q}, \omega)$ obtained for a $\delta = 6.25\%$ hole-doped Hubbard Hamiltonian, again with $V = 0$. (g)(h) $S(\mathbf{q}, \omega)$ for a $\delta = 6.25\%$ hole-doped Hubbard Hamiltonian with an attractive nearest-neighbor interaction $V = -t$. Note that the theory spectra shown in (c)–(h) have been convolved with a Gaussian line shape to reflect the experimental resolution (see Methods). The red circle and arrow in panels (b) and (h) show the features that the Hubbard + V simulations reproduce: a commensurate inelastic peak and a kink in the dispersion (c.f. Fig. 4 below).

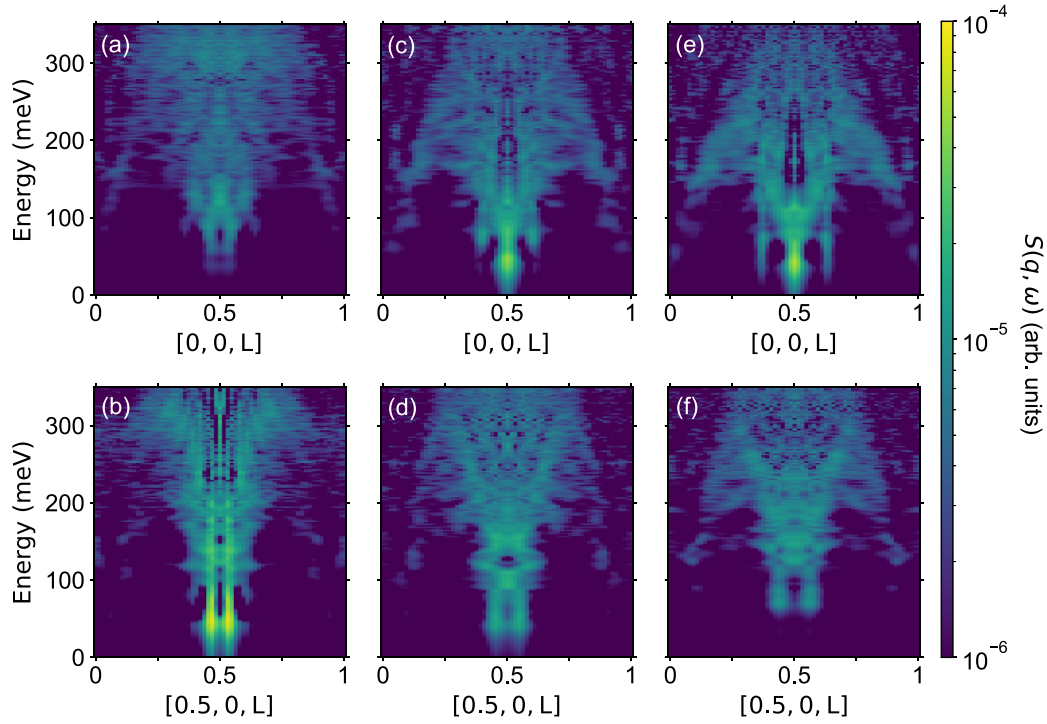


FIG. 3. DMRG simulated dynamical spin structure factor for the doped Hubbard model without extended interactions. The first column [(a) and (b)] shows $S(q, \omega)$ results for 6.25% doping along $q = (q_{\perp}, 0, L)$ with $q_{\perp} = 0$ and π , respectively. The second [(c) and (d)] and third [(e) and (f)] columns show the corresponding dynamical spin structure factor results for 9.375% and 12.5% doping, respectively. All results were obtained on 64×2 site Hubbard ladders.

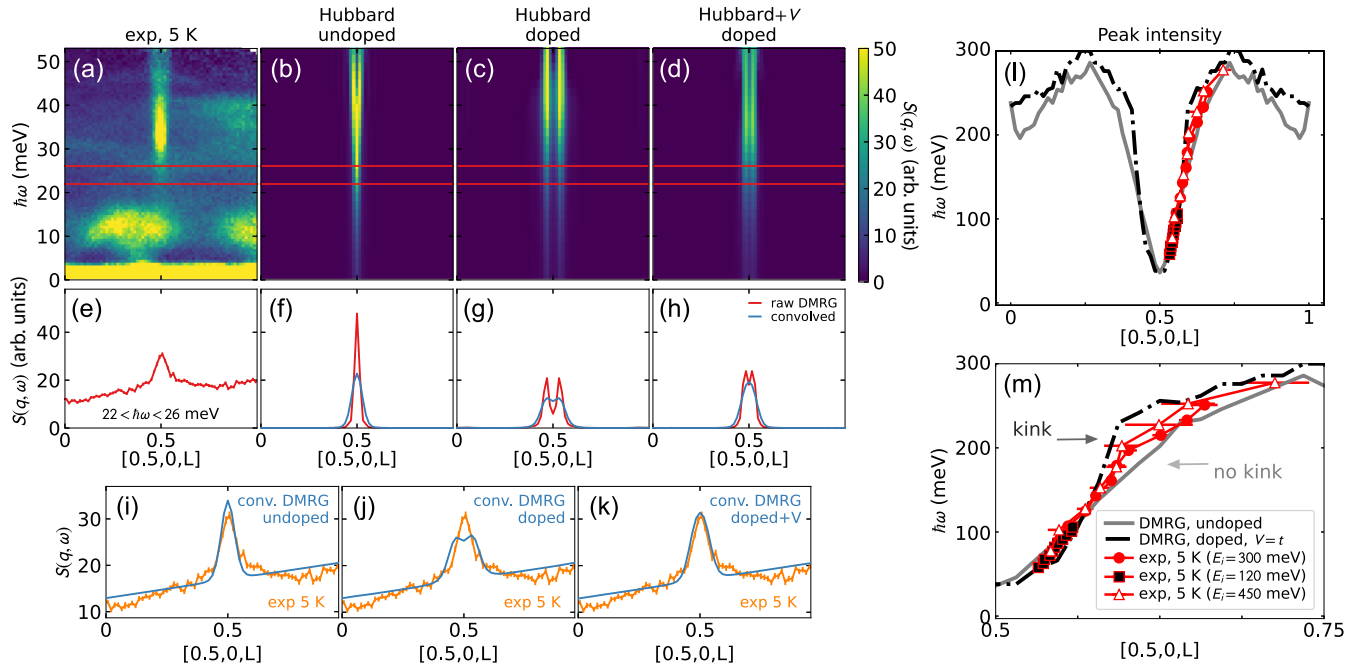


FIG. 4. Lack of incommensuration in the low-energy scattering. (a) The experimental $[\pi, 0, L]$ scattering at 5 K. (b)–(d) DMRG-calculated scattering for the undoped, doped, and extended Hubbard models, respectively. (e)–(h) Cuts through the above panels between 22 meV and 26 meV (shown by the horizontal red lines). According to DMRG simulations, as the doping increases, the peak at $L = \pi$ splits to become incommensurate, but the addition of attractive interactions V causes the peaks to come together again. Convolved with the experimental resolution [blue lines in (i)–(k)], the slightly incommensurate peaks in the Hubbard + V model become a single broad peak. As shown in (i)–(k), the experimental scattering is resolution-limited and commensurate at $L = \pi$, indicating no observable incommensurability. (l)–(m) The fitted dispersion of magnetic excitations for $q_{\perp} = \pi$ (red circles and line). DMRG results for undoped (gray lines) and the doped Hubbard + V model (dotted-dashed lines) are also shown. The kink in the dispersion at $q_{\parallel} = \pi (1 \pm 0.125)$ is reproduced by the doped Hubbard + V model but not the undoped Hubbard model.

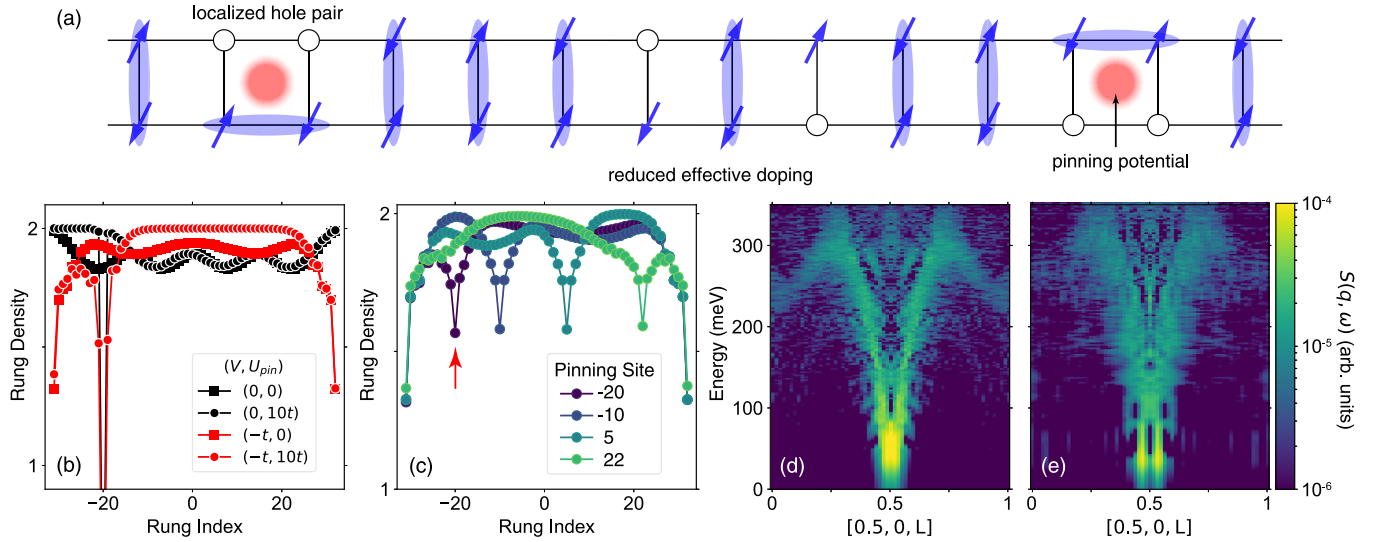


FIG. 5. The combined role of enhanced pairing and impurities in reducing the effective doping. (a) The distribution of doped holes in the ladder. The attractive interaction V enhances hole pairing, increases the effective mass of the carriers, and makes them prone to localization. Pair localization reduces the effective doping of the ladder and minimizes the disruption to the antiferromagnetic correlations. (b) The rung electron density of a 64×2 site Hubbard ladder with a hole pinning potential U_{pin} placed on the 20th rung. The holes are distributed uniformly throughout the cluster when $V = 0$ and $U_{\text{pin}} = 0$. When $V = -t$ but no potential is applied ($U_{\text{pin}} = 0$), the open ends of the cluster act as pinning centers that reduce the carrier concentration in the center. When $V = -t$ and a strong $U_{\text{pin}} = 10t$ is introduced, holes instead collect around both the impurity potential and the open boundaries. Finally, a strong $U_{\text{pin}} = 10t$ without V localizes two holes on the rung. (c) The rung electron densities for an extended Hubbard model with $V = -t$ and a weak $U_{\text{pin}} = t/2$ placed on different rungs. Because of the large value of V , even a weak pinning potential is sufficient to trap many holes near the impurity sites. (d) $S(\mathbf{q}, \omega)$ along $q_{\perp} = \pi$ for one of the cases shown in (c), as indicated by the red arrow. (e) $S(\mathbf{q}, \omega)$ along $q_{\perp} = \pi$ for the $V = 0, U_{\text{pin}} = 10t$ case shown in (b).

and above the charge ordering temperature $T_{\text{CO}} \sim 80$ K [8]), although the 450 meV setting was only measured at 5 K. The complete data set is shown in Fig. 2 within the Supplemental Material [24].

SCCO's magnetic scattering should be independent of the K reciprocal-space direction owing to the decoupled ladder planes. We can thus gather all the relevant information about the magnetic spectrum in the H and L directions from a fixed angle scan. To verify this, we also measured the inelastic spectra at $E_i = 60$ meV while rotating the sample by 180° , and found that the magnetic scattering did not vary with K (see Fig. 3 within the Supplemental Material [24]).

To determine the background, we also measured the scattering in the same configurations (incident energies 30, 60, 120, 300, and 450 meV) with the sample removed. We then smoothed this background data with a Gaussian profile to make up for the smaller counting statistics and subtracted them from the sample scattering, shown in Fig. 4 below. After subtracting the background, we corrected for the anisotropic $d_{x^2-y^2}$ Cu^{2+} form factor [29]

$$f(\mathbf{Q}) = \langle j_0 \rangle + \frac{5}{7}(3 \cos^2 \beta - 1) \langle j_2 \rangle \\ + \frac{3}{56}[35 \cos^4 \beta - 30 \cos^2 \beta \\ + 35 \sin^4 \beta \cos 4\alpha + 3] \langle j_4 \rangle,$$

where β is the angle between $\mathbf{Q}$ and the $d_{x^2-y^2}$ orbital's z axis (the b direction), and α is the xy plane angle measured from the x axis. The $\langle j_n \rangle$ constants for Cu^{2+} were taken from Ref. [30]. In this case, the difference between the isotropic and anisotropic form factors is significant at high energies because

the $d_{x^2-y^2}$ orbitals are ordered in the Cu ladder planes; see Fig. 5 within the Supplemental Material [24]. After subtracting the background and correcting for the form factor [which takes us from intensity to $S(\mathbf{q}, \omega)$], we were able to combine the data from the five different incident energies to produce the plots shown throughout this work. The overall intensities of the spectra taken at different energies were rescaled by comparing constant-energy integrated intensities of the dispersive magnetic modes to ensure that the overall intensity was consistent across the different energy settings.

C. Measured magnetic response

Figures 2(a) and 2(b) show SCCO's measured dynamical spin structure factor $S(\mathbf{q}, \omega)$ at 5 K for $q_{\perp} = 0$ and $q_{\perp} = \pi$, respectively. For an undoped ladder, these spectra probe two- and one-triplon excitations, respectively. The resolution into two parity components modulated by a cosine form factor in $q_{\perp}$ is also evident in the spectra, indicating that parity largely remains a good quantum number for $x = 11.5$.

III. THEORY SIMULATIONS

A. Model

To model SCCO's magnetic response, we performed DMRG simulations of an effective single-band extended Hubbard model defined on a two-leg ladder. The Hamiltonian for

our model is

$$\begin{aligned} \mathcal{H} = & -t_{\parallel} \sum_{l,\mu,\sigma} (c_{l,\mu,\sigma}^{\dagger} c_{l+1,\mu,\sigma} + \text{H.c.}) \\ & -t_{\perp} \sum_{l,\sigma} (c_{l,1,\sigma}^{\dagger} c_{l,2,\sigma} + \text{H.c.}) \\ & -t' \sum_{l,\sigma} (c_{l,1,\sigma}^{\dagger} c_{l+1,2,\sigma} + c_{l,2,\sigma}^{\dagger} c_{l+1,1,\sigma} + \text{H.c.}) \\ & + U \sum_{l,\mu} n_{l,\mu,\uparrow} n_{l,\mu,\downarrow} + V \sum_{\langle l,\mu,l',\mu' \rangle} n_{l,\mu} n_{l',\mu'}, \end{aligned} \quad (1)$$

where $c_{l,\mu,\sigma}^{\dagger}$ ($c_{l,\mu,\sigma}$) creates (annihilates) an electron with spin σ ($=\uparrow, \downarrow$) on rung l and leg index μ ($=1, 2$). The corresponding number operators are $n_{l,\mu,\sigma} = c_{l,\mu,\sigma}^{\dagger} c_{l,\mu,\sigma}$ and $n_{l,\mu} = \sum_{\sigma} n_{l,\mu,\sigma}$. The three different hopping amplitudes distinguish the different bonds of the ladder geometry. They include nearest-neighbor hopping along the legs $t_{\parallel} \equiv t$ and rungs $t_{\perp}$, and diagonal next-nearest-neighbor hopping t' , as indicated in Fig. 1. Throughout this work, we fix $t = 400$ meV, $t_{\perp} = 0.84t$, and $t' = -0.3t$. These parameter choices are identical to those employed in a recent resonant inelastic x-ray scattering (RIXS) study of the parent compound $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ [31]. The interaction parameters U and V denote the on-site and nearest-neighbor density–density interactions, respectively, and are varied while keeping the effective exchange interaction $J = 4t^2/(U - V)$ fixed.

B. DMRG simulations

We calculated the model’s ground- and excited-state properties using zero-temperature DMRG calculations [7,32] performed with the DMRG++ software package [33]. All calculations were performed on long $L \times 2$ ladders with open boundary conditions.

The dynamical spin structure factor $S(\mathbf{q}, \omega)$ is the central quantity characterizing the spin dynamics of ladder materials and constitutes the direct theoretical counterpart of inelastic neutron scattering experiments. It encodes the spatiotemporal spin-spin correlations and describes how the system absorbs energy ω and momentum $\mathbf{q}$ from an external probe. Importantly, $S(\mathbf{q}, \omega)$ can be directly extracted from the inelastic neutron scattering cross section, up to known kinematic factors and the magnetic form factor, making it the experimentally accessible measure of the magnetic excitation spectrum. $S(\mathbf{q}, \omega)$ is given by the Fourier transform of the spin-spin correlation function

$$\begin{aligned} S(\mathbf{q}, \omega) = & \frac{1}{N} \sum_{\substack{\alpha,l,l' \\ \mu,\mu'}} \int_{-\infty}^{\infty} \frac{dt}{2\pi} e^{i[\omega t - \mathbf{q} \cdot (\mathbf{R}_{l,\mu} - \mathbf{R}_{l',\mu'})]} \\ & \times \langle S_{l,\mu}^{\alpha}(t) S_{l',\mu'}^{\alpha}(0) \rangle, \end{aligned} \quad (2)$$

where N is the number of rungs (unit cells) of the ladder, $S_{l,\mu}^{\alpha}$ ($\alpha = x, y, z$) are local spin operators on rung l and leg μ , and $\mathbf{R}_{l,\mu}$ is the position of the site. The spin operators can be expressed as fermionic bilinears in the electron creation and

annihilation operators as

$$S_{l,\mu}^{\alpha} = \frac{1}{2} \sum_{\sigma,\sigma'} c_{l,\mu,\sigma}^{\dagger} \sigma_{\sigma\sigma'}^{\alpha} c_{l,\mu,\sigma'}, \quad (3)$$

where $\alpha = x, y, z$, and σ^{α} are the Pauli matrices acting in spin space. We only compute the $\alpha\alpha = zz$ component because the Hamiltonian is rotationally invariant.

Spin ladders can be viewed as two coupled chains, which naturally leads to a decomposition of the magnetic response into even and odd-rung-parity sectors, corresponding to transverse momenta $q_{\perp} = 0$ and $q_{\perp} = \pi$, respectively. Consequently, $S(\mathbf{q}, \omega)$ provides direct access to the dispersion and spectral weight of the elementary magnetic excitations, such as triplons and multiparticle continua, while explicitly reflecting the ladder geometry and the strength of interleg couplings through its momentum dependence.

Ground-state calculations were carried out on 64×2 ladders, retaining up to $m_{\text{max}} = 2000$ DMRG states. The dynamical spin structure factor $S(\mathbf{q}, \omega)$ was computed using the Krylov correction-vector method [34–36] within the centered-site approximation. For the dynamical calculations, we kept up to $m_{\text{max}} = 1000$ states, employed a Lorentzian energy broadening with half-width at half-maximum $\eta = 0.03t$, and used a frequency step $\Delta\omega = 0.005t$. All DMRG spectra for $S(\mathbf{q}, \omega)$ were uniformly rescaled to match the overall experimental intensity. We also applied a resolution (Gaussian) broadening in momentum and energy to the spectra shown in Figs. 1, 2, and 4(i)–4(k) to facilitate direct comparison with the experimental data. In this case, the momentum and energy broadening were set to match the experimental resolution, with $\sigma_q = 0.237 \times 2\pi$ and $\sigma_{\omega} = 0.055$ meV.

C. Calculated magnetic response

We first compare the measured magnetic response with DMRG calculations for the Hubbard model with $U = 8t$ and without extended interactions ($V = 0$). Figures 2(c) and 2(d) show the calculated $S(\mathbf{q}, \omega)$ obtained using an undoped Hubbard ladder. As discussed in the introduction, SCCO’s ladder subsystem is known to be hole-doped with estimates spanning a relatively wide range, between approximately 6–20% excess holes per Cu (see Ref. [2] and supplementary Fig. 4 of Ref. [13]). Figures 2(e) and 2(f) therefore show results for the same model but obtained for a hole doping $\delta = 0.0625$, corresponding to the lower end of the experimentally inferred range. In both cases, the theory data are plotted along the $\mathbf{q} = (q_{\parallel}, 0)$ (top row) and $(q_{\parallel}, \pi)$ (bottom row) directions, which are equivalent to the $(0, 0, L)$ and $(0.5, 0, L)$ directions in the experiments. Surprisingly, the experimentally measured spectra bear a much closer resemblance to the undoped theoretical results than to those obtained for even this minimal level of hole doping. For example, while the DMRG calculations for the doped case have clear incommensurate magnetic peaks around (π, π) for $\delta = 6.25\%$, no such features are observed in the experimental data. By contrast, the results for the undoped model are much closer to the measured spectra. (There are, however, some notable discrepancies that we will return to momentarily.)

The disagreement between the theory and experiment becomes worse as the doping level increases. To illustrate this,

Fig. 3 summarizes the evolution of the $S(\mathbf{q}, \omega)$ for the Hubbard model ($U = 8t$, $V = 0$) for 6.25, 9.375, and 12.5% hole doping. We find that the low-energy magnetic spectral weight shifts progressively away from the commensurate antiferromagnetic wave vector as the hole concentration increases, leading to a pronounced enhancement of incommensurate spin fluctuations. The systematic evolution of the peak positions with doping is consistent with an approximately linear increase of the incommensurability, in agreement with theoretical expectations for doped single-band Hubbard ladders.

Figure 3 makes it clear that the magnetic response of the SCCO ladder system is inconsistent with that of the doped single-band Hubbard model. This conclusion, however, is at odds with the estimated doping levels of the cuprate ladders obtained from other experiments that are sensitive to the ladder holes. Recent studies on doped one-dimensional (1D) cuprate chains [37,38] and undoped cuprate ladders [31] have concluded that a sizable next-nearest-neighbor attractive interaction $V \approx -t$ is needed to describe the excitation spectra of these materials within single-band frameworks. Motivated by this, we also carried out DMRG simulations for the doped extended Hubbard model with $V = -t$. The results, shown in Figs. 2(g) and 2(h), more closely resemble the experimental data both with regard to the one- and two-triplon excitations. In particular, the splitting of the incommensurate peaks in the triplon dispersion is reduced, implying a reduction in the effective doping, while the spectra are broadened in momentum space from residual charge fluctuations. Below, we will show that these effects are the direct result of increased pairing interactions and Cooper pair localization in the system.

The comparison between experiment and theory shown in Fig. 2 suggests that the SCCO's magnetic response can be captured either by an undoped single-band Hubbard model or by a doped model with a sizable next-nearest-neighbor attraction. We can distinguish between these scenarios by examining the detailed spectra, which reveals several additional and subtle differences between the experiment and predictions of the undoped single-band Hubbard model. Figure 4(a) focuses on the low-energy magnetic response measured along $[0.5, 0, L]$ at 5 K. The corresponding DMRG results for the undoped and doped Hubbard ladders with $V = 0$, and for a doped extended model ($V = -t$) are shown in Figs. 4(b)–4(d), respectively. While doping the Hubbard ladder ($V = 0$) leads to a clear splitting of the magnetic response into incommensurate peaks [Fig. 4(c)], the experimental data remain sharply centered at the commensurate wave vector $q_{\parallel} = \pi$. By contrast, the inclusion of an attractive nearest-neighbor interaction [Fig. 4(d)] suppresses the incommensurate splitting and restores a single broad commensurate peak, in much closer agreement with experiment. This comparison is further clarified by the momentum cuts shown in Figs. 4(e)–4(h), which are obtained by integrating the measured spin response between 22 and 26 meV [indicated by the horizontal lines in Figs. 4(a)–4(d)]. According to the DMRG simulations, increasing the hole doping in the Hubbard model without extended interactions leads to a visible splitting of the peak at $L = 0.5$, reflecting the emergence of incommensurate spin correlations. When an attractive V is included; however, the peaks move back toward commensuration. After convolution with the experimental momentum resolution [Figs. 4(i)–4(k)], the weak residual

incommensuration in the Hubbard + V model can no longer be resolved, resulting in a single peak centered at $q_{\parallel} = \pi$, whose width is consistent with the experimental observations.

Since the precise doping levels of the ladder subsystem are not known at this time, we focus on the lower bound of the experimental estimates for the remainder of this discussion. We have found that increased doping levels can be compensated for by increasing the strength of the attractive interaction V in our analysis. Focusing on a conservative value of 6.25%, therefore, allows us to assess the minimal interaction strength required to suppress the incommensurate response.

In addition to the absence of resolvable incommensurate splitting, the experimental spectra exhibit additional features that are not captured by the undoped Hubbard model. First, the magnetic excitations at $q_{\perp} = \pi$ display a pronounced momentum broadening that exceeds the instrumental resolution and becomes more pronounced at higher energies. Second, the fitted dispersion of the magnetic excitations, shown in Figs. 4(l) and 4(m), has a distinct kink at $q_{\parallel} = \pi(1 \pm 0.125)$ that is absent in the simulations for the undoped Hubbard ladder. Notably, this kink is also absent in measured spectra for the related undoped ladder compound $\text{La}_4\text{Sr}_{10}\text{Cu}_{24}\text{O}_{41}$ [9]. This kink feature is instead well reproduced by the doped extended ($V \neq 0$) Hubbard model, lending further support to this scenario.

Although the overall peak energies of the magnetic response in the experiment ($\Delta_T \approx 32$ meV) and in the DMRG calculations (≈ 38 meV) are comparable, the measured spectra exhibit weak but finite sub-gap scattering extending below the broadened gap edge. Figures 4(a)–4(k) examine this low-energy response in detail. A crucial first step is to distinguish between ladder and chain contributions: the latter appear as a separate band around 10 meV because of their substantially smaller superexchange interaction, and can be identified by the absence of symmetry about $q_{\parallel} = \pi$, reflecting the distinct chain lattice spacing of approximately $c/\sqrt{2}$. The absence of spectral weight below the chain contribution implies that the remaining sub-gap signal retains a finite spin gap $\Delta_{\text{subgap}} \gtrsim 5$ meV.

Because the experimental momentum resolution is finite, it is generally difficult to distinguish a single broadened peak from two closely spaced incommensurate features. However, as shown in Figs. 4(i)–4(k), the resolution-convolved dynamical structure factor obtained from the doped Hubbard model without extended interactions ($V = 0$) retains a clear double-peak structure, allowing us to rule out this scenario. By contrast, the Hubbard + V model, once convolved with the experimental resolution, yields a single broad commensurate peak and provides the best overall agreement with the data. This model also offers an improved description of the higher-energy magnetic excitations at low temperature, correctly capturing both the bandwidth of the triplon dispersion and the characteristic kink near $q_{\parallel} = \pi(1 \pm 0.125)$, as shown in Figs. 4(l) and 4(m).

D. Role of pair localization

The sensitivity of the spin fluctuations to doping originates from the fact that the kinetic energy of the holes is minimized when the spins on neighboring sites along the

leg direction are antiferromagnetically aligned. As a consequence, each doped hole introduces an antiphase boundary in the staggered magnetization favored by the effective antiferromagnetic Heisenberg interactions [39]. This mechanism induces a modulation of the antiferromagnetic correlations with a wavelength set by the average interhole spacing, leading to dominant dynamical spin fluctuations at incommensurate wavevectors $q_{\parallel} = \pi(1 \pm \delta)$. The absence of such incommensurate features in the experimental spectra therefore stands in direct contradiction with the expectations for a doped Hubbard ladder.

A natural route to suppressing these incommensurate correlations is the annihilation of opposite antiphase boundaries, which can be achieved by enhancing the hole-hole attraction. A stronger attraction binds doped holes into more compact Cooper pairs, reducing their coherence length ξ and thereby reinstating dominant commensurate antiferromagnetic fluctuations near $q_{\parallel} = \pi$. In an effective single-band description, such an enhanced pairing tendency can be modeled by introducing a nearest-neighbor attractive interaction $V \approx -t$. As noted earlier, interactions of this magnitude have recently been inferred in doped 1D spin-chain cuprates [37,38] and in the related ladder $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ [31], and have been attributed to strong electron–phonon coupling [37,40]. We also note that a recent DMRG study argued that aspects of the RIXS data could be captured using an effective t - J model [41], which produces stronger pairing correlations relative to the Hubbard model.

Further scrutiny of the DMRG simulations, however, reveals that enhancing pairing alone is insufficient to capture the lack of incommensuration in the doped system. In particular, we also require a degree of carrier (pair) localization induced by pinning centers associated with impurities or disorder [13]. This observation is consistent with the insulating behavior of SCCO at low temperatures, which implies that doped holes are effectively localized at ambient pressures. In the DMRG simulations shown in Figs. 2 and 3, the cluster’s open boundaries act as the pinning centers when $V \neq 0$. However, we can also model these effects by introducing attractive pinning centers at sites throughout the cluster. In both cases, we find that their presence amplifies the role of V by enhancing Cooper-pair localization and reducing ξ . While a small residual density δ' of holes may remain mobile in the bulk, the associated incommensurate peaks at $q_{\parallel} = \pi(1 \pm \delta')$ become correspondingly weak. For sufficiently small δ' , the resulting incommensuration falls below the experimental momentum resolution, naturally explaining the close similarity between the measured spectra and those of the undoped ladder. Taken together, these results indicate that the magnetic response of the doped SCCO family can be understood in a scenario where doped holes experience enhanced Cooper-pair localization.

To test the robustness of this interpretation, Fig. 5 presents DMRG simulations in which pinning centers are explicitly distributed within the ladder, rather than being generated implicitly by open boundary conditions. Figure 5(a) provides a schematic overview of this scenario. Figure 5(b) shows the rung electron densities for a ladder with a strong pinning potential $U_{\text{pin}} = 10t$ placed on a rung on the far left side of the cluster ($l = -20$). When $V = 0$, the pinning center captures only two holes, while the rest remain mobile in the

bulk. The resulting magnetic response, shown in Fig. 5(e), closely resembles that of a conventionally doped Hubbard ladder and is inconsistent with the experimental observations. Moreover, disorder averaging over a distribution of impurities does not resolve this discrepancy [13]. By contrast, when an attractive interaction $V < 0$ is included, tightly bound Cooper pairs accumulate around the pinning center. Crucially, this localization also occurs for weak disorder; Fig. 5(c) shows the rung-resolved charge densities obtained upon introducing substantially weaker attractive pinning potentials $U_{\text{pin}} = t/2$ at different rungs of the ladder. Remarkably, even in this weak-pinning regime, each impurity binds a comparable amount of charge when $V = -t$, demonstrating that the attractive interaction strongly amplifies the impact of weak disorder. The corresponding dynamical spin structure factor $S(\mathbf{q}, \omega)$ along $q_{\perp} = \pi$, shown in Fig. 5(d) for a representative configuration (results for the other configurations are provided in the Appendix, see Fig. 6), confirms that such weak pinning is already sufficient to suppress the incommensurate spin fluctuations.

A direct comparison between Figs. 2(d) and 5(d) highlights that an attractive interaction $V = -t$ is essential for achieving quantitative agreement with the experimental spectra within an effective single-band description of the doped ladders. We have also explicitly verified that the dominant pairing correlations for $V < 0$ possess a d -wave symmetry [1,42], in agreement with recent numerical and analytical studies [20,31,43,44].

Our numerical results show that enhanced pair binding—and the concomitant localization of Cooper pairs—is crucial for reproducing the experimentally observed magnetic spectra within a single-band model. The amplification of localization in this regime can be naturally attributed to the increase in the effective mass of the Cooper pairs associated with the reduced coherence length ξ . Applying external pressure is expected to promote further carrier delocalization, providing a natural explanation for the emergence of superconductivity under pressure in SCCO. More generally, systematic studies of pressure effects and electron–phonon coupling in ladder materials are likely to shed new light on the microscopic mechanisms governing charge pairing and the superconducting transition, including the evolution of the magnetic excitation spectrum and the fate of the incommensurate wavevector upon entering the superconducting state.

IV. SUMMARY AND CONCLUSIONS

We have investigated the magnetic excitation spectrum of doped cuprate ladder compounds using inelastic neutron-scattering measurements and compared the experimental results with large-scale DMRG calculations for single-band Hubbard and extended Hubbard ladder models. Despite substantial hole doping inferred from transport and spectroscopic measurements, the experimental dynamical spin correlations of SCCO remain remarkably close to those of an undoped ladder. In particular, the magnetic response remains commensurate at $q_{\parallel} = \pi$, exhibits only moderate broadening, and shows no evidence of the incommensurate spin fluctuations that are generically expected in uniformly doped ladder systems.

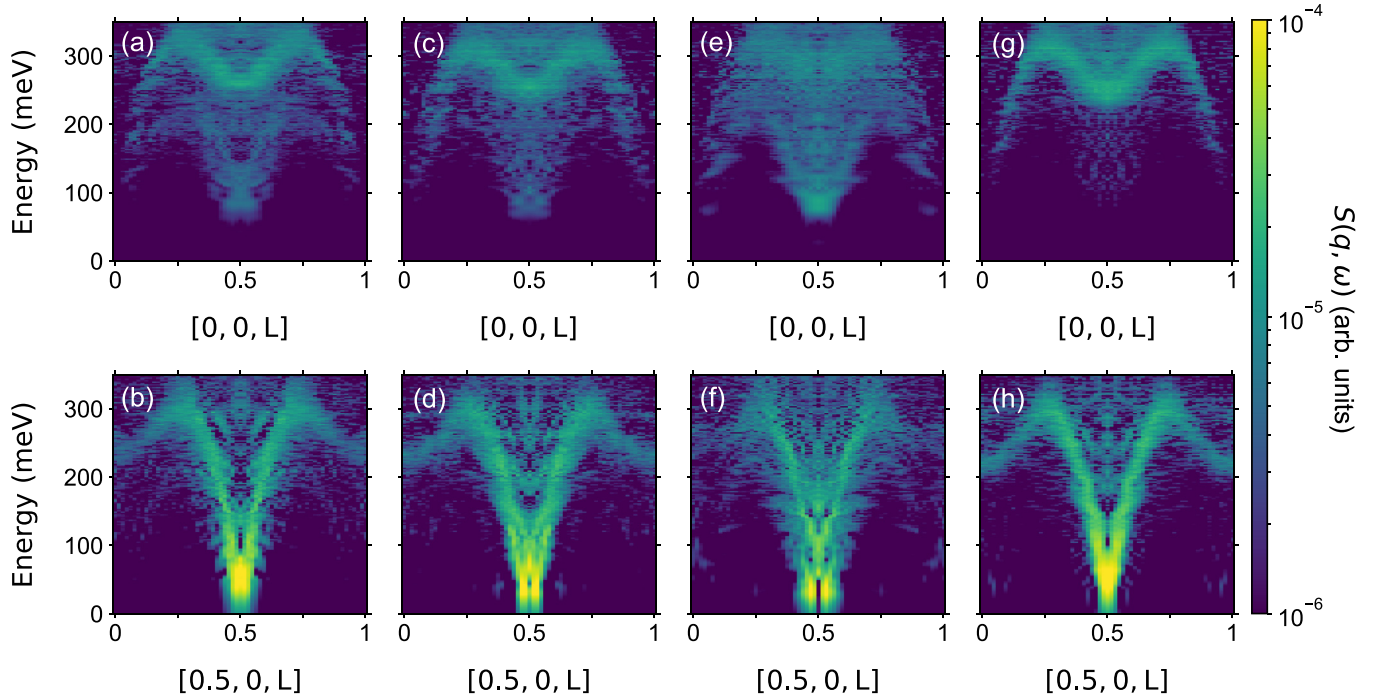


FIG. 6. DMRG-simulated dynamical spin structure factor for the doped extended Hubbard model ($V = -t$) with a weak potential placed on different rungs. The top row shows results for $q_{\perp} = 0$ with a weak potential $U_{\text{pin}} = t/2$ placed on rungs (a) -20 , (c) -10 , (e) 5 , and (g) 22 of the cluster. The bottom row [(b), (d), (f), and (h)] shows the corresponding spectra for $q_{\perp} = \pi$. The main text also reports the case with pinning on rung -20 .

This observation is highly nontrivial in light of the well-established physics of doped single-band Hubbard ladders. In the absence of extended interactions ($V = 0$), hole doping drives the system into a Luther–Emery liquid with a gapless charge mode and gapped spin excitations, where doped holes form bound pairs but simultaneously act as antiphase boundaries in the staggered magnetization. As a result, the magnetic correlations become strongly modulated by the hole density, leading to pronounced incommensurate spin fluctuations whose wave vectors scale linearly with doping. The absence of such features in the INS spectra of doped SCCO therefore places stringent constraints on any effective low-energy description of these materials.

Our DMRG results demonstrate that this discrepancy can be resolved within an effective single-band framework if the Hubbard model is augmented by a substantial attractive nearest-neighbor interaction. The inclusion of such an interaction enhances hole pairing, reduces the coherence length ξ of the Cooper pairs, and increases their effective mass. In the presence of even weak pinning potentials associated with impurities or disorder, this enhanced pairing tendency leads to strong localization of Cooper pairs, effectively reducing the density of mobile carriers in the bulk of the ladder. As a consequence, the incommensurate spin correlations expected for a uniformly doped ladder are strongly suppressed, and the magnetic response remains dominated by commensurate antiferromagnetic fluctuations, in agreement with the experiment. Importantly, disorder alone is insufficient to produce this behavior; localization is substantially amplified only when combined with an attractive interaction of magnitude $|V| \sim t$. A recent DMRG study concluded that the experimental data

can also be understood using an effective t - J model description [41]. This conclusion is consistent with ours in that the t - J model has stronger pairing tendencies than the Hubbard model in the intermediate correlation regime $U/t \approx 8$ that describes the cuprate ladders.

The necessity of introducing a strong attractive nearest-neighbor interaction within a single-band model raises a broader question regarding the adequacy of such a description for cuprate ladder compounds. An alternative interpretation of our results is that a single-band Hubbard model does not capture the correct low-energy degrees of freedom of these materials, and that additional interactions [40] or oxygen orbitals and multiband effects must be explicitly included. Our results thus lead to a clear conclusion: either the low-energy physics of doped cuprate ladders requires a multiband description that goes beyond the single-band Hubbard model, or the effective interactions must be augmented by a sufficiently strong attractive term that eliminates the incommensurate spin fluctuations not observed in the INS data if a single-band framework is retained. In both cases, the experimental magnetic response places tight constraints on the nature of charge dynamics and pairing in these systems.

More broadly, cuprate ladders provide a compelling platform for exploring the interplay of charge pairing, magnetism, and localization in reduced dimensions. Long regarded as stripped-down analogues of high-temperature superconductors, ladder systems additionally highlight the importance of localization effects arising from their composite structure. Within this framework, the pressure-induced superconducting transition observed in SCCO can be naturally interpreted as resulting from increased carrier delocalization under pressure.

Systematic studies of pressure effects, multiband physics, and electron–phonon coupling in ladder materials thus offer promising routes toward elucidating the microscopic mechanisms governing charge pairing, magnetism, and superconductivity in correlated cuprate systems.

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DATA AVAILABILITY

The data supporting this study has been deposited in an open online repository [46].

APPENDIX: RESULTS FOR DIFFERENT IMPURITY CONFIGURATIONS

Figure 6 presents $S(\mathbf{q}, \omega)$ for the four weak-impurity configurations shown in Fig. 5(c). The inferred doping levels are consistent with the evolution of the incommensurate features observed in $S(\mathbf{q}, \omega)$ along $q_{\perp} = \pi$; larger doping concentrations lead to an increased separation between the incommensurate peaks.

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