Unravelling the Nature of the Spin Excitations Disentangled from the Charge Contributions in a Doped Cuprate Superconductor

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The nature of the spin excitations in superconducting cuprates is a key question toward a unified understanding of the cuprate physics from long-range antiferromagnetism to superconductivity. The intense spin excitations in the over-doped regime revealed by resonant inelastic X-ray scattering bring new insights as well as problems—like understanding their persistence or their relation to the collective excitations in ordered magnets (magnons). Here, we study the evolution of the spin excitations upon hole-doping the superconducting cuprate $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8\delta$ by disentangling the spin from the charge excitations in the experimental cross section. We compare our experimental results against density matrix renormalization group calculations for a $t$-$J$-like model on a square lattice. Our results unambiguously confirm the persistence of the spin excitations, which are closely connected to the persistence of short-range magnetic correlations up to high doping. This suggests that the spin excitations in hole-doped cuprates are related to magnons—albeit short-ranged.

I. INTRODUCTION

Starting from antiferromagnetic Mott insulators, the cuprate high-temperature superconductors go through various quantum states with the charge carrier doping as the tuning parameter and form a universal doping-temperature phase diagram. For the hole-doped cuprates, superconductivity emerges in the intermediate doping regime in close proximity to the notorious pseudogap state and the recently established charge density wave state [1]. Disentangling the physics behind these intertwined states is a major challenge for constructing a complete theory of the superconducting cuprates. Fundamentally, the competition between the exchange energy of the localized spins and the kinetic energy of the doped holes is believed to dominate the basic physics, and the two energy scales are naturally regulated by the amount of doped holes [2]. While the holes tend to delocalize and turn the system into a Fermi liquid at high doping levels, electron correlations and local antiferromagnetic correlations survive with modest doping in the superconducting regime and coexist with the well-defined quasiparticles [2–5]. Exactly how these spin and charge degrees of freedom act and interact throughout the doping-temperature phase diagram is therefore a crucial question towards the formulation of a definitive theory of superconductivity in the doped cuprates.

Experimentally, the dynamics and interactions of the spin and charge degrees of freedom are studied by assessing the momentum and energy dependence of the elementary excitations as a function of doping and temperature across the phase diagram. As suggested in numerous papers over the last 11 years [6–27], resonant inelastic X-ray scattering (RIXS) observes intense spin excitations in a large momentum space area around the Brillouin zone center, which persist well into the over-doped region. Crucially the spin excitations in doped samples disperse along the $(\pi, 0)$ direction similarly to the magnons in the antiferromagnetic phase with the damping increasing moderately, although they rapidly become overdamped with doping along the $(\pi, \pi)$ direction and show almost nondispersive profiles [14, 15, 17, 20–23, 26, 27]. This suggests that in some particular area of the Brillouin zone, that is mostly in the $(\pi, 0)$ direction, the magnetic excitations completely ‘ignore’ the existence of a critical value of the doping $\delta$ at which Fermi liquid behavior takes over the correlated magnetism and, moreover, that these excitations resemble the well-known magnons in undoped cuprates (hence their name—paramagnons). Naturally, such results are very much counterintuitive for they lead to an apparent paradox related to the small changes of the spin excitations in the doped cuprates, despite the rapid collapse of the long-range magnetic order upon doping and the dominant Fermi-liquid nature in the over-doped regime. This has sparked an intensive discussion on whether the observed magnetic excitations should indeed be viewed as paramagnons—or rather incoherent particle-hole excitations with a spin-flip [14, 15, 24, 28–30].

To justify the nature of the spin excitations in cuprates as well as the reason of their persistence upon doping, it is necessary to precisely evaluate the momentum and doping evolution of the intrinsic spin excitations and com-
II. RESULTS AND DISCUSSIONS

A. Disentangling the spin and charge excitations in the RIXS spectra

We studied the spin and charge excitations of three different doped Bi2212 samples from under-doped to overdoped regime. The three samples are labeled as UD (Tc = 73 K), OD1 (Tc = 88 K) and OD2 (Tc = 65 K), as shown in Fig 1a. To disentangle the overlapping spin and charge excitations in the Cu L3-edge RIXS spectra, we applied the recently proposed azimuthal dependent method [33], which resolves the two kinds of excitations based on their distinct scattering tensors. In this method, a sample on a wedged sample holder is rotated to change the orientation of the photon polarization in the sample space (see Fig. 1b and Methods section), which gives rise to different rotation dependences according to the scattering tensors [33]. Fig. 1c,d show the azimuthal dependences of the non-spin-flip and spin-flip excitations at \( \sigma \) and \( \pi \) incident polarizations with a 40° wedge angle (\( \theta_w \)). (c) and (d) are the ‘bare’ \( \phi \) dependence on infinite thin samples, while (e) and (f) give the intensity evolution on bulk samples including the self-absorption effect at an energy loss of -0.25 eV.

FIG. 1. Experimental setup and the azimuthal dependence of non-spin-flip and spin-flip RIXS responses. (a) Schematic temperature-doping phase diagram for Bi2212. The purple, green and cyan areas represent the antiferromagnetic, pseudogap and superconducting states respectively. The solid circles indicate the locations of the three measured samples (UD, OD1, OD2) in the phase diagram. (b) The scattering geometry of the azimuthal dependent RIXS experiment on a wedged sample. (c) – (f) The azimuthal \( \phi \) dependence of non-spin-flip and spin-flip RIXS responses in cuprates with a 40° wedge angle (\( \theta_w \)). (c) and (d) are the ‘bare’ \( \phi \) dependence on infinite thin samples, while (e) and (f) give the intensity evolution on bulk samples including the self-absorption effect at an energy loss of -0.25 eV.

prehensively compare to theoretical calculations. This is a difficult task due to the experimental difficulties in extracting \( S(q, \omega) \) from RIXS spectra and also due to the problems in reliably calculating \( S(q, \omega) \). One of the major experimental obstacles comes from the mixing of spin and charge excitations in the RIXS spectra of doped cuprates [31, 32]. With increasing doped holes, one would expect the charge excitations to be stronger, which will worsen the ‘mixing’ problem. This strongly hampers the correct assignment of the spectral profile to solely spin-flip containing excitations, and casts doubts on whether RIXS indeed observes the persistence of the intrinsic spin excitations.

Here we report a systematic study on the momentum and doping evolution of the disentangled intrinsic spin and charge excitations in superconducting Bi2Sr2CaCu2O8+\( \delta \) (Bi2212). By applying the azimuthal-dependent analysis based on the distinct scattering tensors [33], we show that the low-energy excitations of the Cu L3-edge RIXS spectra can be well described by spin-flip (spin) and non-spin-flip (charge) components for all studied doping levels and momenta, which allows us to extract the intrinsic spectral weights of the two components. We find that the spin spectral weight only slightly increases (decreases) with doping at intermediate momentum q along the (\( \pi, \pi \)) [(\( \pi, 0 \))] direction, which unequivocally confirms the persistence of the spin excitations in doped cuprates. We then compare the above experimental results to state-of-the-art density matrix renormalization group (DMRG) calculations of \( t-J \)-like models. The detailed comparison reveals the key characteristics of the spin excitations in the doped cuprates: On one hand, we unravel the crucial role of the longer-range hoppings, as the second- and third-nearest-neighbor hopping are needed to fully reproduce the experimental spin spectrum; on the other hand, we show that solely the short-range magnetic correlations are enough to qualitatively reproduce the persistence of the intensity of the spin excitations upon doping. Our results thus suggest that RIXS indeed observes the persistent spin excitations in hole-doped cuprates and that their paramagnetic nature can be understood as stemming from localised spins with short-range correlations.
in the low-energy RIXS spectra.

Fig. 2a and b show the RIXS intensity map of OD2 sample at \( q = (0.33, 0) \) as a function of azimuthal angle \( \phi \) and energy loss \( E \) at \( \sigma \) and \( \pi \) incident polarizations, respectively. As the energies of \( d-d \) excitations are situated well above 1 eV \([14, 15, 34]\), it is natural to assume that the low-energy excitations are mainly composed of the single-spin-flip and non-spin-flip excitations involving the \( 3d_{x^2-y^2} \) orbital. Although the double spin flip with a net spin change of zero (bimagnon with \( \Delta S = 0 \)) is also allowed in RIXS process in cuprates \([25, 35, 36]\), earlier studies show that its spectral weight quickly diminishes with hole-doping \([37-39]\), and becomes negligible in Cu \( L_3 \) RIXS when doping is beyond 0.08 \([25]\), which is the case of this study. In addition, the bimagnon intensity in Cu \( L_3 \) RIXS is maximal at the zone center, and becomes significantly weaker at large momentum \([25]\). The low-energy RIXS intensity at a certain momentum \( q \) is thus expressed as a linear combination of the spectral weights of the single-spin-flip and non-spin-flip components modified by their corresponding azimuthal dependence:

\[
I_{RIXS}(E, \phi, \epsilon_i) = w_s(E) \cdot A_s(E, \phi, \epsilon_i) + w_c(E) \cdot A_c(E, \phi, \epsilon_i)
\]

(1)

where \( w_s(E) \) is the spectral weight of spin (charge) components, and \( A_{s(c)}(E, \phi, \epsilon_i) \) is the azimuthal dependence which is already known from the scattering tensor and self-absorption effects, and \( \epsilon_i \) is the incident polarization. Using the azimuthal dependent RIXS intensity, the spin and charge spectral weights can be obtained by solving the above system of linear equations. Fig. 2c-f show the \( \phi \) dependence of RIXS intensity at constant energy loss of 0 and -0.25 eV with the decomposed spin and charge contributions \( w_s(E) \cdot A_{s(c)}(\phi) \). As can be seen, the RIXS azimuthal dependences can be well fitted by these two components, which verifies that the above analysis correctly describes the low-energy RIXS response in Bi2212. In addition, the quasi-elastic scattering at 0 energy loss is dominated by charge-like \( \phi \) dependence, consistent with the charge nature of the quasi-elastic peak. In Fig. 2g,h, we compare the RIXS spectra at two special geometries, grazing incidence \( (\phi = 0^\circ) \) with \( \sigma \) polarization and grazing emission \( (\phi = 180^\circ) \) with \( \pi \) polarization, with the decomposed spin and charge components. The grazing emission with \( \pi \) polarization is usually used to measure the magnetic excitations in cuprates, since the charge component is largely suppressed in this geometry as shown in Fig. 2h. Nonetheless, the charge component is still considerable, which could influence the correct evaluation of the profile and intensity of magnetic excitations. It is therefore necessary to fully disentangle the spin and charge components to precisely study their nature. We note here that the obtained spectral functions \( w_{s(c)}(E) \) are solely related to the properties of the studied samples, as the angle and polarization related geometry factors and the self-absorption effect are removed by the knowledge of \( A_{s(c)}(E, \phi, \epsilon_i) \). This allows the direct and unambiguous comparison between \( w_{s(c)}(E) \) and theoretical calculations based on different models, which could provide vital knowledge to understand the spin and charge excitations in cuprates.

Fig. 3 presents the obtained spin and charge spectral functions \( w_{s(c)}(E) \) of the three different doped samples at different momenta \( q \) along both the \((\pi, 0)\) and \((\pi, \pi)\) directions. Fig. 3a-g show the decomposed spin spectral functions, which all show a single peak with a damped profile. Fig. 3h plots the energy integrated intensity \( I_s(q) \) of the spin spectral functions. The \( I_s(q) \) of all three samples show a similar \( q \) dependence: \( I_s(q) \) monotonically increases with increasing \( q \), and has a slightly larger intensity when approaching large \( q \) along \((\pi, \pi)\) direction.
FIG. 3. Momentum and doping dependence of the decomposed spin and charge spectral functions. (a) – (g) ((i) – (o)) display the spin (charge) spectral functions for samples of different doping. The solid lines in (a) – (g) indicate the fittings by a damped harmonic oscillator model. (h) Energy integrated intensities of the spin spectral functions. (p) Energy integrated intensities of the charge spectral functions with the quasi-elastic peaks subtracted.

In contrast to the spin response, this charge response shows much stronger intensity increases along (π, 0) direction while it saturates around (0.25, 0.25) along (π, π) direction. These excitations are likely the electron-hole continuum. We note that previous SGA+1/N calculations show that the charge-dynamic susceptibility of the electron-hole continuum has larger intensities along (π, 0) direction than (π, π) direction [40], which is consistent with the results here.

By comparing different doping levels, one can notice that the spin excitations show different development along the (π, 0) and (π, π) directions: the total spectral

than (π, 0) direction. This q dependence is qualitatively consistent with the results in a previous study which calibrate the geometry influences by comparing to the INS results [27]. Fig. 3i-o show the decomposed charge response at different q. There are two main components in the charge response: a quasi-elastic peak with low-energy phonons close to zero energy loss, and a broad peak around -0.4 eV which extends to high energy loss. The quasi-elastic peak is enhanced at (0.13, 0.13), which is due to the structure modulation at (0.125, 0.125) in Bi2212 samples. Fig. 3p shows the integrated intensity of the broad peak after the quasi-elastic peak is subtracted.
weight increases with increasing doping at intermediate $q$ along $(\pi, \pi)$, while it slightly decreases along $(\pi, 0)$ direction, as shown by $I_s(q)$ in Fig. 3h. On the other hand, the spectral weights of the decomposed charge excitations simply increase with increasing doping along both directions, while the increase is more remarkable along $(\pi, 0)$ direction than $(\pi, \pi)$ as shown in Fig. 3p. A previous RIXS study on single layer (Bi,Pb)$_2$(Sr,La)$_2$CuO$_{6+\delta}$ [26] also investigated the influence of doping on the spin excitations using the grazing-emission and incident $\pi$-polarization geometry which enhances the scattering contribution from the spin-flip channel. In contrast, they found the intensity of spin excitations increases with doping at small and intermediate $q$ along both $(\pi, 0)$ and $(\pi, \pi)$ directions, but crosses over to decrease at large $q$, although the increase becomes very subtle along the $(\pi, 0)$ direction when the doping level enters the superconducting regime. These different results could originate from either the differences between single and double layer cuprates, or a small residual mixture with charge excitations in the grazing-emission and incident $\pi$-polarization geometry. Note that the $(\pi, 0)$ direction will endure more influences from the residual charge excitations as the charge excitations are more intense and doping-dependent along the $(\pi, 0)$ direction (see Fig. 3p).

The distinct spin excitations response to the hole doping along $(\pi, 0)$ and $(\pi, \pi)$ directions could provide important information for the understanding of the spin dynamics in doped cuprates, which can be obtained by comparing with state-of-the-art theoretical calculations discussed in detail in the next part of the paper.

For a more quantitative analysis of the spin response, we fit the spin spectral functions $\omega_s(q)(E)$ by a damped harmonic oscillator (DHO) model convoluted with a resolution function (see Methods). As shown by the solid lines in Fig. 3a–g, the results can be well fitted by the DHO model. Fig. 4 presents the fitting results. The bare frequency $\omega_0$ is similar for all three dopings, while the damping $\gamma$ increases with increasing doping and has a much larger value along $(\pi, \pi)$ direction. This is consistent with previous studies on doped cuprates showing that the magnetic excitations are much more damped along $(\pi, \pi)$ direction [14, 15, 17, 20–23, 26, 27]. With the charge excitations excluded, we can now rule out that the over-damped profile of the spin excitations along $(\pi, \pi)$ direction comes from an increase of charge contributions with doping. We note that the fitted damping factors of the two smallest momenta of OD2 sample are large and out of the main trend of the momentum dependence. We attribute this to the decomposed shape of spin excitations bearing more influences from the uncertainties in the azimuthal fitting when the spin excitation peak is getting closer to the elastic peak at small momenta. Fig. 4b shows the propagation frequency defined as $\omega_p = \sqrt{\omega_0^2 - \gamma^2}$, where a zero value is assigned when the system is over damped, i.e. $\omega_0 < \gamma$. One can see that the spin excitations in the over-doped OD2 sample are fully over damped along the $(\pi, \pi)$ direction. Fig. 4c shows the fitted proportional amplitude $A$ to DHO model. It increases with increasing doping along $(\pi, \pi)$ direction while it changes little along $(\pi, 0)$ direction, which is a bit different from the integrated total spectral weight $I_s(q)$ shown in Fig. 3h. This is mostly due to $I_s(q)$ including both the effects from the proportional amplitude $A$ and the damping $\gamma$, while $A$ excludes the effect of damping $\gamma$ which suppresses the total intensity.

**B. Unravelling the character of the spin excitations in doped cuprates**

The model of choice to study the evolution of the spin excitations upon doping the cuprates is the “celebrated” $t$–$J$–$t'$-like model [41], defined by the Hamiltonian on a 2D square lattice:

$$H = -t \sum_{\langle i,j \rangle} (\tilde{c}_i^\dagger \tilde{c}_j + h.c.) - t' \sum_{\langle\langle i,j \rangle\rangle} (\tilde{c}_i^\dagger \tilde{c}_j + h.c.) - t'' \sum_{\langle\langle i,j \rangle\rangle} (\tilde{c}_i^\dagger \tilde{c}_j + h.c.) + J \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} \tilde{n}_i \tilde{n}_j),$$

where $\tilde{c}_i^\dagger$ operator creates an electron at site $i$ in the constrained Hilbert space without double electron occupancies, $\mathbf{S}_i$ is a spin-1/2 operator at site $i$ and $\tilde{n}_i$ is the on-site electron density at site $i$: $\tilde{n}_i = \tilde{c}_i^\dagger \tilde{c}_i$. The model parameters $t$, $t'$ and $t''$ denote the hopping integrals between first, second and third neighbors, respectively, whereas $J$ is the antiferromagnetic Heisenberg interaction between nearest neighbor spins. In our calculations, we take a realistic (and widely-accepted [42]) choice [43] of the values...
of the $t$-$t'$-$t''$-$J$ model parameters $t' = -0.3t$, $t'' = 0.15t$ and $J = 0.4t$; besides, to better understand the role of the longer-range hoppings, we also consider switching off the $t''$ hopping (the $t$-$t'$-$J$ model) or both the $t''$ and $t'$ hoppings (the $t$-$J$ model). Note that, while to describe electronic properties of the cuprates probably the charge-transfer $(pd)$ model would be more appropriate, the spin excitations are believed to be well-described by models with oxygens being integrated out [26, 31, 44–48]. The $t$-$t'$-$t''$-$J$ model on a square lattice has been intensively studied as a host of superconducting state for a long time (for example, see [49, 50] and references therein), and a possible existence of superconducting phase in a wide range of hole doping has been proposed [51].

Next our goal here is to compute the static spin structure factor $S(q)$, typically defined as:

$$S(q) = \langle S(-q) \cdot S(q) \rangle = \frac{1}{N} \sum_{i,j} \langle S_i \cdot S_j \rangle e^{i(q \cdot r_i - q \cdot r_j)},$$

where the $i$, $j$ indices run over all sites, $N$ is the number of sites and $r_i$ denotes the position of the site in the lattice. This is done by involving state-of-the-art DMRG calculations which are carried out on an $N = 6 \times 6$ square lattice with open boundary conditions (OBC). We chose a $6 \times 6$ OBC cluster to investigate a wide range of parameters with high accuracy, see Supplementary. The use of OBC enables us to avoid an artificial enhancement of specific period correlations, which frequently occurs in periodic and cylindrical systems. However, due to the OBC, the charges tend to localise at the edges when charge imbalance, i.e. holes, is introduced. To counterbalance this effect, we introduce an edge factor $\lambda$ that multiplies the electronic hopping parameters $t$, $t'$ and $t''$ as well as the spin exchange coupling $J$ acting on the sites on the perimeter of our cluster, see Methods.

By using the real space approach we have full control over which contributions to $S(q)$ we include in Equation 3. In fact, we can select the distance $\ell$ at which the sum in Equation 3 is truncated. In particular, it is possible to consider the different total averages for the different spin-spin correlations $\langle S_i \cdot S_j \rangle_\ell$ with $\ell = 1, 2, 3, \ldots$ defining the considered neighbors. The difference in $S(q)$ between using the singular values of the spin-spin correlations and the averages is minimal (see Supplementary). While shielding us from accessing $S(q, \omega)$, this approach allows us to thoroughly study the possible magnonic character of the persistent spin excitations.

We now discuss how our theoretical calculations compare to the experimental results presented in Fig. 3. Our aim within this calculations is to reproduce the switching in the sequence of intensities upon doping. Hence, our focus is on this qualitative aspect of the experimental results, rather than on the quantitative reproduction of the experimental data within our theoretical calculations. The main results are shown in Fig. 5: whereas Fig. 5a presents the experimental integrated intensity of spin spectral weights for the three measured doping levels UD, OD1 and OD2 (see above), Fig. 5b shows the calculated $S(q)$ with the $t$-$t'$-$t''$-$J$ model in the restricted Brillouin zone kinematically available to the experiments. The crucial message here is that, overall, the qualitative agreement between theory and experiment is solid. First, the experimentally observed surprisingly small anisotropy between the $(\pi, \pi)$ and $(\pi, 0)$ directions is also reproduced by our calculations—albeit it is not as small as in the RIXS experiment. Second, at five crucial momentum points the theoretical calculations give the same sequence of intensities of the spin structure factor as a function of doping as the experiment (see five grey circles in Fig. 5b). In detail, the calculations reproduce: (i) the small increase in intensity upon doping at small $q$ in the $(\pi, 0)$ direction, (ii) the large increase in intensity upon doping observed at moderate $q$ in the $(\pi, \pi)$ direction, (iii) the decrease in intensity upon doping observed at large $q$ in the $(\pi, 0)$ direction, and finally (iv) almost no changes in intensity upon doping observed at large $q$ in the $(\pi, \pi)$ direction.

The agreement between the $t$-$t'$-$t''$-$J$ model and experiments can be appreciated even more after looking at Fig. 6, where we present the comparison between the theoretical results of the three different $t$-$J$-like models. Fig. 6a shows $S(q)$ calculated for the ‘bare’ model. Within this model, the sequence of intensities of the spin structure factor as a function of doping does not change
as a function of momentum. It means that the dependence of the static spin structure factor on momentum does not alter upon doping, unlike observed in the experiments. Fig. 6b shows the same quantity as Fig. 6a, but for the $t$-$t'$-$J$ model. In this case, results in the $(\pi, \pi)$ direction show the same qualitative behavior as in the experimental case. However, one important feature is missing in the $(\pi, 0)$ direction, namely the $S(q)$ of the lowest doped system, $n = 0.11$, does not 'overtake' the static structure factor of the higher doped systems at large $q$, large referring to the largest momenta kinematically available to the experiments. In order to achieve our goal, i.e., the correct sequence of intensities of the spin static structure factor as a function of doping and momentum along the $(\pi, 0)$ direction, we need to include the third neighbor hoppings. Once the $t$-$t'$-$t''$-$J$ model is considered, we are able to reproduce most of the experimentally observed behavior, as shown in Fig. 5. Based on these results, the importance of long range hopping becomes apparent.

Due to our real space approach, we have full control over the contribution of spin correlations of different range to the static spin structure factor. In fact, we can cut the summation in Equation 5 (Equation 6 for $n = 0.22$) to include only up to a certain number of neighbors. This analysis allows us to investigate to which degree magnetic correlations contribute upon doping. In Fig. 7, we show the main results of this analysis on the model that is able to qualitatively reproduce the experimental results, namely the $t$-$t'$-$t''$-$J$ model. Fig. 7a is a cartoon description of the main real space spin-spin correlations needed in our $6 \times 6$ cluster in order to qualitatively reproduce the persistence of the intensity of the spin excitations upon doping. The color scale represents the value of such real space correlations and we considered one sample site around the centre of the cluster and its neighbors up to third-neighbors. In Fig. 7b we plot the result of cutting the sum in Equation 5 (Equation 6 for $n = 0.22$) to include only up to third-neighbors spin-spin correlations. While a small gap appears around $q = (0, 0)$, the results qualitatively agree with the full Fourier transform depicted in Fig. 6c. In particular, the sequence of intensities of the spin static structure factor for the different doping levels is the same at five momen-

FIG. 6. **Theoretical static spin structure factor and the longer-range electronic hoppings.** Static spin structure factor $S(q)$ obtained using DMRG on a $6 \times 6$ cluster (see text for further details) for three different hole-doping levels and (a) the $t$-$J$ model, (b) the $t$-$t'$-$J$, and (c) the $t$-$t'$-$t''$-$J$ model. Model parameters as in Fig. 5. Note the enlarged momentum coverage w.r.t. Fig. 5 and different scales of $S(q)$ for the $(\pi, \pi)$ and $(\pi, 0)$ directions of the Brillouin zone.

FIG. 7. **Theoretical static spin structure factor and the short-range magnetic correlations.** (a) The $6 \times 6$ cluster used in the DMRG calculations and the nearest neighbor (NN), next nearest neighbor (NNN) and third neighbor (3rd N) spin bonds; the color scale applied to the sites shows the value of the average spin-spin correlation for those particular neighbors. (b) Static spin structure factor $S(q)$ obtained using DMRG on a $6 \times 6$ cluster (see text for further details) for three different hole-doping levels and for the $t$-$t'$-$t''$-$J$ model (parameters as in Fig. 5) with keeping only first, second and third neighbor spin-spin correlations in the Fourier Transform of Eq. (4). Note the enlarged momentum coverage w.r.t. Fig. 5 and different scales of $S(q)$ for the $(\pi, \pi)$ and $(\pi, 0)$ directions of the Brillouin zone.
tum points in both the $(\pi, \pi)$ and $(\pi, 0)$ directions as observed in the experiments. This means that considering short-range magnetic correlations in $t$-$J$ like models is enough to recover the main properties of the spin static structure factor in the $(\pi, \pi)$ and $(\pi, 0)$ directions, as also recently suggested for the Hubbard model in [52]. Moreover, the appearance of this artificial gap at $q = (0,0)$ can be understood by noticing that, by including only short-range real space correlations, we look at large-$q$ values. Indeed, by adding longer-range correlations, the artificial gap disappears. An in depth discussion of these properties as well as similar analysis for the $t$-$J$ and $t$-$t'$-$J$ models and the development of the features in the $S(q)$ lines upon including neighbors from first to third can be found in the Supplementary.

III. CONCLUSION

Our main experimental result is the unambiguous assessment of the momentum and doping evolution of the disentangled intrinsic spin and charge excitations measured by Cu $L_3$-edge RIXS in doped Bi2212 samples. By applying the azimuthal dependent cross section analysis based on the scattering tensors, we find that the low-energy part of the RIXS spectra can be well explained as a linear combination of the spin-flip and non-spin-flip responses for a range of dopings and momenta. This allows a precise extraction of the spin and charge spectral weights as functions of momentum and doping without mixing. The extracted charge responses show relatively broad profiles, and their momentum and doping dependences qualitatively agree with the electron-hole-pair excitations which is an important contribution in RIXS cross sections [15, 40]. The spin response, on the other hand, shows profiles which can be well fitted by a damped harmonic oscillator model, although they become over damped along $(\pi, \pi)$ direction. Moreover, the momentum and doping dependence of the spin and charge responses are different, implying the distinct nature of the two responses. The integrated spin excitations spectral weights only change slightly upon doping, with no abrupt reduction in the over-doped sample. As the charge components are exactly separated from the spin, this result confirms that RIXS indeed observes the persistence of spin excitations in a large part of the Brillouin zone in doped cuprates.

The obtained experimental results on the spin excitations are qualitatively reproduced by numerical simulations of the $t$-$J$-like model. It turns out that the bare $t$-$J$ model is not enough and instead this model has to be supplemented by longer-range hoppings—which points out the decisive role of such hoppings in reproducing the experimentally observed paramagnons in doped cuprates. Furthermore, the extensive real space analysis shows that short-range magnetic correlations are needed in order to cause the observed persistence of spin excitations in doped cuprates, meaning they need to be paramagnonic in nature. From that, two important consequences follow: On one hand, within the class of cuprate models with localised spins, those without any magnetic correlations at all seem not to be realistic for doped cuprates. On the other hand, this means that longer-range magnetic correlations do not play a crucial role in the doped cuprates. Altogether, this helps in resolving the paradox related to the persistence of the spin excitations upon doping the cuprates—despite a rapid collapse of the long-range magnetic correlations.

On the theory side, there are three important implications of the results presented in this paper: The first one is that this work shows that a recent theoretical suggestion that the spin excitations are responsible for the $T$-linear dependence of the electronic scattering in the Hubbard model [52] might indeed become a realistic scenario for the cuprates. This follows from the above-stated conclusion that, without any ambiguities, the collective spin excitations persist in the doped cuprates. The second one follows from the suggested crucial role played by the longer-range hoppings in the $t$-$J$ models. Such a result goes in line with, inter alia, recent works advocating for the strong sensitivity of the phase diagram of the $t$-$J$-like models to the value of the next-nearest neighbor hopping $t'$ [53, 54] and thus ‘sweetens the bad news’ coming from the study suggesting the lack of superconductivity in the ground state of the 2D Hubbard model without longer-range hoppings [55]. The third point relates to the fact that, as stated above, solely the short-range magnetic correlations are needed to explain the persistence of the intensity of the paramagnons in doped cuprates, similar to the recently established well-defined magnons in the random $t$-$J$ model up to 33% hole doping [56]. Thus, an interesting task for theory would be to gain an intuitive understanding of why the short-range magnetic correlation alone can lead to the lack of changes of the paramagnons along the $(\pi, 0)$ direction.

IV. METHODS

A. RIXS Experiments and samples

The RIXS experiments were carried out with the SAXES spectrometer at the ADRESS beamline of the Swiss Light Source at the Paul Scherrer Institut [57, 58]. The incident X-ray energy was set at the Cu $L_3$-edge RIXS in doped Bi2212 samples. By applying the azimuthal dependent cross section analysis based on the scattering tensors, we find that the low-energy part of the RIXS spectra can be well explained as a linear combination of the spin-flip and non-spin-flip responses for a range of dopings and momenta. This allows a precise extraction of the spin and charge spectral weights as functions of momentum and doping without mixing. The extracted charge responses show relatively broad profiles, and their momentum and doping dependences qualitatively agree with the electron-hole-pair excitations which is an important contribution in RIXS cross sections [15, 40]. The spin response, on the other hand, shows profiles which can be well fitted by a damped harmonic oscillator model, although they become over damped along $(\pi, \pi)$ direction. Moreover, the momentum and doping dependence of the spin and charge responses are different, implying the distinct nature of the two responses. The integrated spin excitations spectral weights only change slightly upon doping, with no abrupt reduction in the over-doped sample. As the charge components are exactly separated from the spin, this result confirms that RIXS indeed observes the persistence of spin excitations in a large part of the Brillouin zone in doped cuprates.

The obtained experimental results on the spin excitations are qualitatively reproduced by numerical simulations of the $t$-$J$-like model. It turns out that the bare $t$-$J$ model is not enough and instead this model has to be supplemented by longer-range hoppings—which points out the decisive role of such hoppings in reproducing the experimentally observed paramagnons in doped cuprates. Furthermore, the extensive real space analysis shows that short-range magnetic correlations are needed in order to cause the observed persistence of spin excitations in doped cuprates, meaning they need to be paramagnonic in nature. From that, two important consequences follow: On one hand, within the class of cuprate models with localised spins, those without any magnetic correlations at all seem not to be realistic for doped cuprates. On the other hand, this means that longer-range magnetic correlations do not play a crucial role in the doped cuprates. Altogether, this helps in resolving the paradox related to the persistence of the spin excitations upon doping the cuprates—despite a rapid collapse of the long-range magnetic correlations.

On the theory side, there are three important implications of the results presented in this paper: The first one is that this work shows that a recent theoretical suggestion that the spin excitations are responsible for the $T$-linear dependence of the electronic scattering in the Hubbard model [52] might indeed become a realistic scenario for the cuprates. This follows from the above-stated conclusion that, without any ambiguities, the collective spin excitations persist in the doped cuprates. The second one follows from the suggested crucial role played by the longer-range hoppings in the $t$-$J$ models. Such a result goes in line with, inter alia, recent works advocating for the strong sensitivity of the phase diagram of the $t$-$J$-like models to the value of the next-nearest neighbor hopping $t'$ [53, 54] and thus ‘sweetens the bad news’ coming from the study suggesting the lack of superconductivity in the ground state of the 2D Hubbard model without longer-range hoppings [55]. The third point relates to the fact that, as stated above, solely the short-range magnetic correlations are needed to explain the persistence of the intensity of the paramagnons in doped cuprates, similar to the recently established well-defined magnons in the random $t$-$J$ model up to 33% hole doping [56]. Thus, an interesting task for theory would be to gain an intuitive understanding of why the short-range magnetic correlation alone can lead to the lack of changes of the paramagnons along the $(\pi, 0)$ direction.

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B. Azimuthal dependent RIXS measurements

In two-dimensional superconducting cuprates, the single-spin-flip and non-spin-flip excitations in the 3d_{x^2-y^2} orbital as well as the other d-d excitations in the Cu L3-edge RIXS spectra show distinct geometry and polarization related characters, which are determined by their different scattering tensors [35, 60, 61]. This allows to assess the nature of these excitations by either resolving the polarizations of both the incident and scattered photons [62] or by azimuthal dependent RIXS measurements [33]. In 1b, we show the scattering geometry and the sample rotation of the azimuthal dependent experiments. The directions of the incident and emitted x-rays are fixed through the scattering angle to 130°, thus the total momentum transfer is fixed at \( q \). Two linear polarizations (\( \sigma \) and \( \pi \)) are used for the incident x-rays while the polarization of the emitted light is not resolved. The plate-like sample is mounted on a wedged sample holder (with wedge angle \( \theta_w = 10°, 20°, 40° \) and 50° in the experiments) to have a certain in-plane momentum transfer. The azimuthal rotation axis is parallel to the total momentum transfer. The azimuthal rotation axis is parallel to the total momentum transfer \( q \), so that the projections of \( q \) in the sample reciprocal frame are unchanged during rotation, while the projections of the photon polarization are changing. This allows measuring the azimuthal dependence of the excitations at fixed momentum in the sample momentum space. When rotating the sample, the photon polarization will be continuously rotated in the sample space, and the scattering tensors will then result in different rotation dependences for different excitations.

C. Fitting by damped harmonic oscillator model

The formula of the damped harmonic oscillator (DHO) model used for the fitting of the spin spectral functions in Fig. 3 is:

\[
A \cdot \frac{\gamma \omega}{(\omega^2 - \omega_0^2)^2 + 4\gamma^2 \omega^2} \cdot (4)\
\]

A Gaussian resolution function with 100 meV FWHM is convoluted with the above DHO model to fit the results.

D. DMRG calculations

In numerical calculations with OBC cluster, a proper correction is often added into the Hamiltonian to minimize the effects of missing terms at open edge. In this study, we introduced the edge factor to uniformize the mobility of charge. The correct edge factor is calculated for each doping level and each different model (\( t-t^\prime-J \), \( t-t^\prime-J, t-t^\prime-t^\prime\prime-J \)). We extrapolate it by computing the dispersion \( \delta = n_{\text{in}} - n_{\text{out}} \) where \( n_{\text{in}} \) is the averaged electron density taken over the sites which do not belong to the edges and \( n_{\text{out}} \) is the averaged electron density taken over the sites which form the edge of the cluster. We computed the dispersion \( \delta \) for different values of the edge factor \( \lambda \) for each doping level and model and take the final value of the edge factor \( \lambda \) as that at which the dispersion \( \delta = 0 \). Nevertheless, the obtained values (\( \lambda \sim 0.9 - 1.2 \)) are close enough to 1 to smoothly connect the inside and edge of the cluster. Furthermore, by introducing this edge factor the Friedel oscillations as well as the most important finite-size effect coming from using OBC can be significantly reduced, as can be seen in the supplementary.

We keep up to \( m = 7000 \) states in the DMRG calculations, leading to an error \( \epsilon/N = 10^{-6} \). We make sure that the local density in the system is isotropic by using the edge factor \( \lambda \) as described above and we compute the real space spin-spin correlations \( \langle \vec{S}_i \cdot \vec{S}_j \rangle \) for all pairs \((i, j)\) labelling the system’s sites. This way, we can compute the spin static factor \( S(q) \) as:

\[
S(q) = \sum_{i,j} \langle \vec{S}_i \cdot \vec{S}_j \rangle \cos(q_x(x_i - x_j) + q_y(y_i - y_j)). \tag{5}\n\]

This method is only valid when the local z-component of the spins is small enough, e.g. \( S_i^z \leq 10^{-5} \) for all sites \( i \). However, for certain doping levels and models, it was not possible to reach this level of accuracy for the local value of \( S^z \). Therefore, we renormalized the \( S^z S^z \) correlations and computed \( S^z(q) \) as:

\[
S^z(q) = \sum_{i,j} \langle (S_i^z S_j^z - \langle S_i^z \rangle \langle S_j^z \rangle) \cos(q_x(x_i - x_j) + q_y(y_i - y_j)) \rangle. \tag{6}\n\]

Due to the symmetries of the considered models, \( S(q) = 3S^z(q) \). Equation 6 was used for the doping level \( n = 0.22 \) for both the \( t-t^\prime-J \) and the \( t-t^\prime-t^\prime\prime-J \) models (see Fig. 6).

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reduce the photon penetration depth when grazing occurrence, which will reduce the proportionality especially at the resonance where $\mu$ is large. Such a saturation effect can be described by the following relation [64–66]:

$$ TEY(E, \alpha, \epsilon) \propto \frac{ML}{\lambda(E, \epsilon) \cdot \sin \alpha} \cdot \left(1 + \frac{1}{\lambda(E, \epsilon) \cdot \sin \alpha} \right). $$

where $M$ is a material dependent factor and the second factor on right side expresses the saturation effect. One can see that this saturation factor will distort the shape of $\text{TEY}$ when (1) $L/|\lambda(E, \epsilon) \cdot \sin \alpha| \sim 1$ and (2) the $\lambda(E, \epsilon)$ is strongly energy dependent such as around the absorption edge. To address the possible saturation effect in our samples, we measured the TEY at several different incident angles at $\sigma$ polarization, as shown in Fig. 8(a)-(c). The saturation effect is relatively small with only a slight intensity reduction on the resonance peak in the UD and OD1 samples, but becomes more obvious in the OD2 sample. Fig. 8(d) shows the incident angle dependence of $\text{TEY} \cdot \sin \alpha$ at the resonance peak and pre-edge background. The solid lines are the fitting by a function of $A/\left(1 + 1/(r \cdot \sin \alpha)\right)$, in which $r$ gives an estimate of $\lambda/L$. The fitted values of $r$ are 22, 40, and 11 for the UD, OD1 and OD2 sample respectively. We note that this is just a rough estimate due to the limited points of measured angles. Nevertheless, these values qualitatively agree with the previous measurement in YBCO film with $r \sim 20$ [66]. The results suggest that the TEY at normal incidence is nearly proportional to the absorption coefficient. So we take the TEY at normal incidence and $\sigma$ polarization as the in-plane absorption tensor element $f_{aa}(E)$. For the TEY at $\alpha = 20^\circ$ and $\pi$ incidence, some saturation effects exist. However, as the resonance peak amplitude is relatively small ($\sim 15\%$) comparing to the pre-edge background, the saturation factor is much less energy dependent comparing to the $\sigma$ polarization. Thus, we can use an overall $E$-independent correction factor determined by the change of the pre-edge background from $\alpha = 90^\circ$ to $\alpha = 20^\circ$ at $\sigma$ polarization to correct the spectra. By subtracting the contribution of $\sin^22\theta f_{aa}(E)$, one can then get the out-of-plane element $f_{cc}(E)$. Fig. 8(e) shows the obtained $f_{aa}(E)$ and $f_{cc}(E)$ of all three samples, which are normalized to make the pre-edge background the same. As can be seen, there are almost no resonance peaks in $f_{cc}(E)$, which is due to the in-plane $3d_{x^2-y^2}$ character of the samples. $f_{aa}(E)$ and $f_{cc}(E)$ are almost the same for the three samples, except a small intensity reduction in the resonance peak of $f_{aa}(E)$ in OD2 sample. As the doped holes mainly goes to the oxygen ligands, this main peak should be mostly unaffected while the shoulder at a bit higher energy ($\sim 934.2$ eV) increases with the hole doping [67, 68]. The reduction is thus most likely due to some residual saturation effects in normal incidence in OD2 sample due to its smaller $\lambda/L$ ratio at resonance. In our analysis, we use the same set of $f_{aa}(E)$ and $f_{cc}(E)$ from OD1 sample for all the three samples.

The self-absorption effect in RIXS is described by the

**Appendix A: Absorption coefficients and self-absorption correction**

Self-absorption is always present in soft X-ray RIXS measurements on bulk samples. It will largely modify the RIXS intensity and has to be considered in azimuthal dependent measurement. To derive its contribution, one needs the knowledge of the linear absorption coefficient $\mu(E, \epsilon)$ of the studied sample, which depends on the energy $(E)$ of the X-rays as well as the orientation of the polarization vector $(\epsilon)$ in the sample space. For a single crystal, the absorption coefficient can be expressed in a tensor form, which is constrained by the point group symmetry of the crystal. The Bi2212 sample has a tetragonal structure, leading to a diagonal absorption tensor with only two independent elements, $f_{aa}(E)$ and $f_{cc}(E)$, which correspond to the absorption coefficients with polarization vector in the sample $a$-$b$ plane and along the out-of-plane $c$ axis, respectively [63]. The absorption coefficient with arbitrary polarization vector is,

$$ \mu(E, \epsilon) = \epsilon^T \cdot \begin{pmatrix} f_{aa}(E) & 0 & 0 \\ 0 & f_{aa}(E) & 0 \\ 0 & 0 & f_{cc}(E) \end{pmatrix} \cdot \epsilon. $$

(A1)

In this study, we use the X-ray absorption spectroscopy (XAS) measured by total electron yield (TEY) to evaluate the absorption tensor elements $f_{aa}(E)$ and $f_{cc}(E)$ of the Bi2212 samples around the Cu L3-edge. The inset of Fig. 8 displays the geometry of the measurements. The in-plane element $f_{aa}(E)$ is probed by $\sigma$ polarized X-ray, while the out-of-plane element $f_{cc}(E)$ is probed at grazing incidence and $\pi$ polarization. As the fully grazing incidence is not possible, here we use $20^\circ$ incident angle, which gives a result of $\sin^22\theta f_{aa}(E) + \cos^22\theta f_{cc}(E)$. The TEY signal is usually proportional to the absorption coefficient giving that the electron escape depth $L$ is much smaller than the photon penetration depth $\lambda \cdot \sin \alpha$, where $\lambda = 1/\mu$ and $\alpha$ is the incident angle [64–66]. Although $L \ll \lambda$ is usually true in cuprates [66], $\sin \alpha$ will reduce the photon penetration depth when $\alpha$ is small at
The absorption tensor elements $f_{aa}(E)$ and $f_{cc}(E)$ for three samples can be expressed as a product of the intrinsic spectral types of excitations $(e)$ and $(f)$ for the non-spin-flip and spin-flip excitations at an energy loss of -0.25 eV are shown in Fig. 8 precisely, and we can therefore calculate the self-absorption tensor $\Sigma(E, \varphi, \epsilon)$ for our samples, with the knowledge of the absorption tensor of the studied excitation and a rotation matrix which links the laboratory coordinates and sample coordinates, as already demonstrated in reference [33].

$$I_{t}^{\exp}(E, \varphi, \epsilon_{i}) = \sum_{\epsilon_{f}} I_{t}(E, \varphi, \epsilon_{i}, \epsilon_{f}) = \sum_{\epsilon_{f}} I_{t}(E, \varphi, \epsilon_{i}, \epsilon_{f}) = \sum_{\epsilon_{f}} I_{t}(E, \varphi, \epsilon_{i}, \epsilon_{f})$$

Here the $I_{t}^{\exp}(E, \varphi, \epsilon_{i})$ is the experimental (exp) RIXS intensity for a certain type $(t)$ of excitation which can be spin, charge or $d$-$d$ excitations, and $I_{t}(E, \varphi, \epsilon_{i}, \epsilon_{f})$ is the intrinsic RIXS intensity with $E = E_{f} - E_{t}$ the energy loss. The denominator describes the self-absorption effect, where $\mu(E_{t})$ is the absorption coefficient of the incident (emitting) photons, $\hat{k}_{f}(\varphi)$ is the unit vector of the photon propagation direction, and $\hat{n}(\varphi)$ is the unit vector normal to the sample surface. As the final polarization $(\epsilon_{f})$ is not resolved in this experiment, the measured intensity is a sum of all the possible $\epsilon_{f}$ ($\sigma$ and $\pi$ polarizations). The azimuthal $\varphi$ dependence of $I_{t}(E, \varphi, \epsilon_{i}, \epsilon_{f})$ can be calculated based on the scattering tensor of the studied excitation and a rotation matrix which links the laboratory coordinates and sample coordinates, as already demonstrated in reference [33].

Fig. 1(c), (d) in the main text show the $\varphi$ dependence of $\sum_{\epsilon_{f}} I_{t}(E, \varphi, \epsilon_{i}, \epsilon_{f})$ for the non-spin-flip and spin flip excitations. With the knowledge of the absorption tensor of our samples, $\mu(E_{t})$ can be determined precisely, and we can therefore calculate the self-absorption effect and include it in the final azimuthal dependence $I_{t}^{\exp}(E, \varphi, \epsilon_{i})$ for a certain type of excitation. The results at an energy loss of -0.25 eV are shown in Fig. 1(e) and (f) for the non-spin-flip and spin flip excitations. The final RIXS spectrum is a sum of all possible types of excitations $\sum_{t} I_{t}^{\exp}(E, \varphi, \epsilon_{i})$, and $I_{t}^{\exp}(E, \varphi, \epsilon_{i})$ can be expressed as a product of the intrinsic spectral weight of the excitation $u_{t}(E)$ and the azimuthal dependent geometry factor $A_{t}(E, \varphi, \epsilon_{i})$. In the low-energy part of spectra in cuprates, the spin-flip and non-spin-flip excitations related to $3d_{x^2-r^2}$ orbital dominates, the RIXS intensity is then explained by equation 1, which can be decomposed into the two components by their azimuthal dependence.

### Appendix B: Detailed discussions of the influence of the short-range magnetic correlations and longer-range electronic hoppings on the static spin structure factor

In what follows we study the impact of including solely a restricted number of short range spin-spin correlations in the Fourier transform defining the static spin structure factor $S(q)$, see Eq. (5) in the main text of the paper. The main results are presented in Fig. 9 which shows $S(q)$ calculated in the three distinct $t$-$J$–like models considered in this paper and when up to the third neighbor spin-spin correlations are considered in the Fourier transform. (In the main text, we discuss the case of the $t$-$t'$-$t''$-$J$ model when up to the third neighbor spin-spin correlations are considered.)

First of all, let us look at the results shown in the first column of Fig. 9, i.e. once solely spin-spin correlations up to first-neighbors are included (by definition this includes also on site spin-spin correlations) when calculating $S(q)$. Except for small quantitative differences, the three considered versions of the $t$-$J$ model show the same behavior, meaning the effect of the longer range hopping $t'$ and $t''$ is minimal when the cut-off in the Fourier transform is on the nearest-neighbor correlations. This can be understood when one realises that first neighbors correlations are always relatively large and antiferromagnetic and the subtle effects induced by the longer range hopping terms is minimal. Moreover, due to the fact that the considered correlations are antiferromagnetic (i.e. negative), a negative spectral weight sets in for small $q$ in $S(q)$. We would like to underline that this result is not linked to the instability of the ground state, as also the ground state in these calculations is still the exact ground state of each considered model; instead, this is due to approximate calculations of $S(q)$, namely the respective cuts in the Fourier series (see above). As a side remark let us note that when one considers up to first neighbors in $S(q)$, its behavior is qualitatively the same in the $(\pi, \pi)$ and $(\pi, 0)$ directions of the Brillouin zone. In particular, the damping of the peak at $(\pi, \pi)$ is recovered, but the same behavior happens at $(\pi, 0)$.

Next, we move on to the second column of Fig. 9, where we consider up to second-neighbor spin-spin correlations in the Fourier transform for $S(q)$. Now the two directions in the Brillouin zone, $(\pi, \pi)$ and $(\pi, 0)$, show different behaviors. The damping of the $(\pi, \pi)$ peak is more prominent and the intensity at this point is increased for all three models. No negative weight is present. The
main features present in the $S(q)$ are already present for the $t$-$J$ model. A distinct behavior is now visible when longer range hoppings are included: in the $(\pi, 0)$ direction the different doping lines do not cross nor overlap, while they do so once the “bare” $t$-$J$ model is considered. In the $(\pi, \pi)$ direction, both the $t$-$t'$-$J$ and $t$-$t'$-$t''$-$J$ models already show the onset of a line crossing around $(\pi/2, \pi/2)$. However, as previously stated, no crossing is present in the $(\pi, 0)$ direction, meaning we need to include further neighbor spin correlations in the Fourier transform to recover this behavior.

Finally, we focus on the third column of Fig. 9, where up to third-neighbor spin-spin correlations are included in the Fourier transform. When considering the simpler $t$-$J$ model, there is again an onset of negative weight, which has disappeared in the previous case. This comes from the inclusion of third-neighbor spin-spin correlations, which are antiferromagnetic, i.e., negative. Being quite strong, they are not compensated by the ferromagnetic, i.e., positive, second-neighbors correlations, which leads effectively to a negative weight. On the other hand, the intensity of the $(\pi, \pi)$ peak is now doubled compared to the one seen when only first-neighbor correlations are included. Qualitatively, there is not a big difference if compared to the previous case where up to second-neighbors correlations are being considered. If longer range hoppings are included, the negative weight problem is solved. Let us now focus on the $t$-$t'$-$J$ case: in the $(\pi, \pi)$ direction, the intensity of the peak has increased and it is comparable with that of the full $S(q)$. Furthermore, the crossing at $(\pi/2, \pi/2)$ is present. If we now look at the $(\pi, 0)$ direction, we see the onset of a crossing close to $(\pi, 0)$. Compared to the full $S(q)$ case [see Fig. 6(b)], the 0.11 doping level is now “meeting” the other doping level lines at $(\pi, 0)$. The last case is the one which includes also the $t''$ hopping as already shown in the main text. The behavior in the $(\pi, \pi)$ direction is similar to that seen in the $t$-$t'$-$J$ model just discussed, therefore we will only examine what happens in the $(\pi, 0)$ direction. Here the crossing close to $(\pi, 0)$ is clearly visible and includes all three doping levels, as seen in experiments and in the full $S(q)$.

Lastly, we would like to comment on the presence of a gap at $(0, 0)$ momentum transfer observed in the results for both the second and third column of Fig. 9: it is clear that when one considers the long range distances in real spaces, this translates to small $q$ values in $q$-space. Therefore, the “wrong” behavior seen at small $q$ is ex-

**FIG. 9. Detailed contributions of the short-range magnetic correlations and the longer-range electronic hoppings to the theoretical static spin structure factor** Static spin structure factor $S(q)$ obtained using DMRG on a $6 \times 6$ cluster (see text for further details) for the $t$-$J$ (top panels), the $t$-$t'$-$J$ (middle panels), and $t$-$t'$-$t''$-$J$ model (bottom panels). In all cases only a restricted number of short range spin-spin correlations is nonzero in the Fourier Transform of Eq. (5): solely first neighbors (left panels); solely first and second neighbors (middle panels); and solely first, second, and third neighbors (right panels). Model parameters as in Fig. 5. Note the enlarged momentum coverage w.r.t. Fig 5 and different scales of $S(q)$ for the $(\pi, \pi)$ and $(\pi, 0)$ directions of the Brillouin zone.
expected to improve more and more when further neighbors are included.

In conclusion, in order to understand the behavior of the integrated intensity $S(q)$, it is important to include longer range hopping up to third neighbors, but only shorter range spin-spin correlations are needed to recover the main properties seen in the experimental data.

**Appendix C:** Benchmarking the use of the edge factor in the numerical method

In order to perform calculations on an open cluster, we have introduced the edge factor $\lambda$ as described in Sec. 1. To prove that this method leads to correct results, we calculated the static spin structure factor $S(q)$ on the half-filled $6 \times 6$ cluster. The results are shown in Fig. 10 and fully agree with the textbook behavior of the Heisenberg model on a square lattice at zero temperature [a dominant peak at $(\pi, \pi)$].

Moreover, as mentioned in the main text, using open clusters could provide much better results than periodic clusters if the open edge is managed properly. To confirm this statement, we compare the static spin structure factor $S(q)$ between short and long open chains in the 1D $t$-$J$ chain with $J/t = 0.4$ and $n = 2/3$, since the result for large 2D system is not available. In Fig. 11, the spin static structure factors for $L = 6$ and $L = 60$ open chains are compared. The result for $L = 60$ is expected to be almost identical to taking the thermodynamic limit. For the $L = 6$ open cluster, small local potentials $0.15$ and $-0.3$ have been added on the first and second edge sites, respectively, as edge factors used to achieve a uniform density distribution, i.e., $< n_i > = 2/3$ for all $i$, $i$ labeling the sites in the chain. We find a good agreement between $S(q)$ with $L = 6$ and $L = 60$ except for the peak height around $q = 2\pi/3$. This discrepancy is caused mainly by an enhanced finite-size effect due to strong quantum fluctuations in 1D. Thus, such discrepancy will be much smaller in 2D systems. The structure factor for an $L = 6$ periodic chain is also plotted. It is difficult to compare it with the thermodynamic limit result, because only discrete momenta are allowed. Besides, the artificial enhancement (suppression) of $S(q)$ at $q = 2\pi/3$ (at $q \neq 2\pi/3$) is clearly seen. Consequently, we can suggest that the use of an (small) open cluster is a reasonable way to capture the overall features of $S(q)$, unless $S(q)$ shows a very complex behavior.

**Appendix D:** Averaged compared to non-averaged correlations

In the main text as well as in Appendix B, the results presented are based on the Fourier transform of the averaged values of the different neighbor spin-spin correlations. To support our choice of showing results based on averaged correlations, Fig. 12 compares the spin static structure factor $S(q)$ calculated using the Fourier transform with keeping up to third neighbor spin-spin correlations for the $t$-$t'$-$t''$-$J$ model—with averaged (over the whole cluster) and non-averaged values of the spin-spin correlations. The differences between the two latter cases are almost invisible, suggesting that our choice of consid-
FIG. 12. Theoretical static spin structure factor including averaged and non-averaged short-range magnetic correlations. Static spin structure factor $S(q)$ obtained using DMRG on a $6 \times 6$ cluster for the doped $t$-$t'$-$t''$-$J$ model with only up to third neighbor spin-spin correlations included in the Fourier transform defining $S(q)$. Top (bottom) panels shows results obtained using an averaged (non-averaged) values of the short magnetic correlations, respectively. Model parameters as in Fig. 5. Note the enlarged momentum coverage w.r.t. Fig 5 and different scales of the OY axes for the $(\pi, \pi)$ and $(\pi, 0)$ directions of the Brillouin zones.