Anisotropic damping of the spin fluctuations in doped La$_{2-x}$Sr$_x$CuO$_4$ studied by resonant inelastic x-ray scattering

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We report high-resolution resonant inelastic x-ray scattering (RIXS) measurements of the collective spin fluctuations in three compositions of the superconducting cuprate system La$_{2-x}$Sr$_x$CuO$_4$. We have mapped out the excitations throughout much of the 2-D $(h,k)$ Brillouin zone. The spin fluctuations in La$_{2-x}$Sr$_x$CuO$_4$ are found to be fairly well-described by a damped harmonic oscillator model, thus our data allows us to determine the full wavevector dependence of the damping parameter. This parameter increases with doping and is largest along the $(h,k)$ line, where it is peaked near $(0.2,0.2)$. We have used a new procedure to determine the absolute wavevector-dependent susceptibility for the doped compositions La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.12, 0.16$) by normalising our data to La$_2$CuO$_4$ measurements made with inelastic neutron scattering (INS). We find that the evolution with doping of the intensity of high-energy excitations measured by RIXS and INS is consistent. For the doped compositions, the wavevector-dependent susceptibility is much larger at $(\frac{1}{2},0)$ than at $(\frac{1}{2},\frac{1}{2})$. Thus, the strongest magnetic excitations, and those predicted to favour superconductive pairing, occur towards the $(\frac{1}{2},\frac{1}{2})$ position as observed by INS.

I. INTRODUCTION

The origin of high temperature superconductivity (HTS) in doped layered cuprate materials remains a subject of intense interest in both experimental and theoretical research, despite over 30 years of activity. It is widely believed that the magnetic degrees of freedom and in particular spin fluctuations are primarily responsible for superconductive pairing in the cuprates. 1-4 In this case, it is important to characterise the collective spin excitations as a function of wavevector, energy, doping and temperature to see how they correlate with the occurrence of superconductivity and compare with theoretical models.

Resonant inelastic x-ray scattering (RIXS) 5-11 and inelastic neutron scattering (INS) 15-21 are complementary probes which directly yield information about the wavevector and energy of the dynamical structure factor $S(Q,\omega)$ or dynamic susceptibility (response function) $\chi''(Q,\omega)$ at high frequencies. The La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) system allows the evolution of $S(Q,\omega)$ to be measured across the phase diagram, from the antiferromagnetic (AF) parent compound La$_2$CuO$_4$ (LCO) through superconducting compositions.

In La$_2$CuO$_4$, the spin waves have their lowest energies at the $\Gamma$, $Q = (0,0)$ and $M$, $Q = (\frac{1}{2},\frac{1}{2})$ positions and $\chi''(Q,\omega)$ is small near $\Gamma$ and largest near $M$. INS measurements 13-14 throughout the Brillouin zone have shown that the magnetic excitations can be fairly well-described as spin waves derived from a Heisenberg model with next-nearest neighbour interactions including a ring exchange. As expected, they are strongest near the AF wavevector $Q = (\frac{1}{2},\frac{1}{2})$ and show anomalously strong damping at the $X$ or $(\frac{1}{2},0)$ position. 12,20,22

For superconducting compositions in LSCO, INS shows that the strongest response 15-18 occurs near $Q = (\frac{1}{2},\frac{1}{2})$ at low and intermediate energies ($0-150$ meV), with comparable intensity to the parent antiferromagnet. For optimally doped ($x = 0.16$) LSCO, an incommensurate structure is observed 23 for $h\omega \lesssim 25$ meV. Above 50 meV the magnetic excitations disperse 21-25 away from $(\frac{1}{2},\frac{1}{2})$. At high energies, $h\omega \approx 250$ meV, excitations are observed 25 on the Brillouin zone boundary at $Q = (\frac{1}{2},0)$ in LSCO ($x = 0.14$) demonstrating the persistence of high energy spin excitations for superconducting compositions. For overdoped compositions, 21-23 $x = 0.22 - 0.25$, the lower energy ($h \approx 50$ meV) features observed at optimal doping are suppressed.

Cu $L_3$ RIXS 19-20 measurements of the spin fluctuation in LSCO are complementary to INS. They are restricted to a circular region in $(h,k)$ centered on $\Gamma$ [see Fig. 1 (a) and (b)] but are able to isolate high energy excitations ($h\omega \gtrsim 300$ meV) more easily. Early RIXS measurements in LSCO 19 showed the existence of dispersing spin fluctuations. Spin excitations are observed 19-20 throughout the first AF Brillouin zone including at the boundary [e.g. $(\frac{1}{2},0)$ position] where INS 19 also finds excitations. RIXS studies suggest that these excitations show wavevector-dependent damping 19-20. Spin fluctuations persist to overdoped compositions and evolve relatively slowly with doping 19-20.

The improved energy resolution of the measurements
we have performed allows us to model the nature of the spin fluctuations more precisely. The motivation of this work is to perform a systematic characterisation of the spin fluctuations in LSCO with this enhanced energy resolution including mapping the $Q$-dependence of the frequency and damping throughout a 2-D portion of the Brillouin zone. We also aim to bridge the techniques of INS and RIXS to establish an estimate of the absolute spin susceptibility.

Here we report RIXS measurements on three dopings of LSCO, $x = 0$, 0.12 and 0.16. We have made use of the high resolution and high intensity of the RIXS spectrometers ID32 at the European Synchrotron Radiation Facility (ESRF) and I21 at the Diamond Light Source (DLS) to map out magnetic spectra over 2-D ($h, k$) space. We find that, for doped compositions, the magnetic response is fairly well-described by a damped harmonic oscillator line shape. The pole frequency and damping are strongly anisotropic in agreement with previous studies along the $(h, 0)$ and $(h, h)$ lines, with the strongest damping along the $(h, h)$ line and centred near $(0.2, 0.2)$ for the optimally doped composition. By comparing data on $\text{La}_2\text{CuO}_4$, where the spin waves are well studied, with LSCO, we make quantitative estimates of the wavevector-dependent susceptibility $\chi(Q)$. This quantity is a vital input to theories of the HTS phenomenon\cite{2,3}.
to the beam. The sample used for measurements of the $x = 0.12$ compound at the ESRF (used only in the 2-D map plots) was polished following the procedure in Croft et al.

### B. Notation

LSCO undergoes a structural transition to a low-temperature orthorhombic (LTO) phase below $T_{\text{LTO}} \approx 240$ K, however, we use the high-temperature tetragonal (HTT) $I\bar{4}m\bar{m}$ crystal structure notation to allow comparison between the three compounds. In this notation, $a = b \approx 3.8$ Å, $c \approx 13.2$ Å. The momentum transfer $Q$ is defined in reciprocal lattice units (r.l.u.) as $Q = h\alpha + k\beta + l\gamma$ where $\alpha = 2\pi/a$ etc. The measured excitations are labelled via their energies $\hbar\omega = c|\mathbf{k}| - c|\mathbf{k}'|$ and momenta $Q = \mathbf{k} - \mathbf{k}'$, where $\mathbf{k}$ and $\mathbf{k}'$ are in initial and final wavevectors.

### C. Spectrometers

High resolution RIXS spectra were measured at beamline ID32 of the ESRF [28,29] and the I21 RIXS spectrometer at DLS [30]. The incoming beam energy was tuned to the Cu L$_3$-edge ($\sim 932$ eV) with linear horizontal (LH) $\pi$ polarisation. We present LH data from the grazing-out orientation where the single magnon intensity is favoured [21,22]. Recent experiments with polarisation analysis [33] have established that this configuration is primarily sensitive to magnetic scattering. Samples were mounted on the sample holder in ultra-high vacuum and cooled to $T \approx 20$ K. Magnetic excitations in doped cuprates are dispersive predominantly in the $a$-$b$ plane of LSCO, allowing paths to be measured in the $(h,k)$ plane by varying the sample orientation, and keeping the scattering angle $2\theta$ fixed at $146^\circ$ and $149.5^\circ$ for I21 and ID32 respectively. The scattering geometry is shown in Fig. 1 (a). We assume there is negligible dispersion in the features of interest from variation of $l$, and therefore we focus only on the momentum transferred in the $(h,k)$ plane. Spectra were principally measured along the two high-symmetry lines $(h,0)$ and $(h,h)$ as indicated with red arrows in Fig. 1 (b) with energy resolution $\Delta E \approx 35$ meV. The $x = 0$ and 0.12 measurements were performed at I21 and the $x = 0.16$ measurements were performed at ID32 and repeated at I21. In both doped compounds, further measurements were performed at ID32 with $\Delta E \approx 50$ meV on a grid of $Q$-points evenly distributed throughout a quadrant of the Brillouin zone indicated by the red shaded region in Fig. 1 (b). The energy resolution was established using elastic scattering from a silver paint or carbon tape reference. For I21, a background was measured from either a dark image taken after the collection or by fitting a linear background outside the excitation range, $\leq -0.1$ eV and $\geq 5$ eV.

### D. Analysis

#### 1. Data processing

In order to carry out a quantitative analysis of the data, we follow recent practice [31,32,33,34] and assume that the magnetic intensity observed in RIXS is proportional to the spin-spin dynamical structure factor $S(Q,\omega)$ which is used to interpret neutron scattering experiments [35]. $S(Q,\omega)$ is, in turn, proportional to $\chi'^2(Q,\omega)$ multiplied by the Bose factor $n(\omega) + 1 = [1 - \exp(-\hbar\omega/k_B T)]^{-1}$. Clearly, the scattering processes in RIXS and INS are very different, with the observed RIXS intensity being dependent on the relative orientation of the photon electric field to the Cu 3$d$ orbitals as well as the absorption of the x-ray photons within the sample. These factors are known to vary slowly with $Q$ [36,37]. Nevertheless, to correct for these effects we initially normalise our raw counts $I_{\text{raw}}$ to the energy-integrated dd excitation intensity obtained from the same spectrum. The intensity of the dd excitations is known to be dependent on the polarisation $\epsilon$ and wavevector $\mathbf{k}$ and can be described by a function $g(\epsilon,\epsilon',\mathbf{k},\mathbf{k}')$. We denote the measured intensity $I_{\text{RIXS}}$ as $I_{\text{raw}}/g$ where $g = \int g(\epsilon,\epsilon',\mathbf{k},\mathbf{k}') d\omega$ is the integral described above evaluated over the range $1 - 3$ eV.

The spectra were aligned to the elastic reference and the exact zero-energy position was established by fitting an elastic peak with a Gaussian function. The aligned spectra were modelled within a range $-80$ to $800$ meV. As well as the spin excitations and elastic peak we fitted low-energy excitations, which are interpreted as phonons, using Gaussian functions. Electron-hole excitations contribute to RIXS scattering for $\hbar\omega > 0$ for doped compositions. This contribution was modelled with a constant multiplied by a $n(\omega) + 1$ Bose function, where the width in energy was determined by the instrumental resolution rather than temperature. For the parent compound, multimagnon excitations are resolvable at $\sim 400$–$600$ meV, and are fitted using an additional damped harmonic oscillator pole (see Sec. 11,13).

#### 2. Damped harmonic oscillator model

A damped harmonic oscillator (DHO) model may be used to describe a given spin-wave mode with wave vector $Q$. This approach has recently been taken in a number of RIXS studies [11,13,14,15]. The analogous mechanical DHO equation is [36]

$$\ddot{x} + \omega_0^2 x + \gamma \dot{x} = f/m,$$  \hspace{1cm} (1)

where $\omega_0$ is the frequency of the undamped mode and $\gamma$ is the damping parameter. In our case, both of these are $Q$-dependent, thus $\omega_0 = \omega_0(Q)$ and $\gamma = \gamma(Q)$.

The imaginary part of the response function for a given
wavevector can be written as,

$$
\chi''(Q, \omega) = \frac{\chi'(Q) \omega_0^2(Q) \gamma(Q) \omega}{\omega^2 - \omega_0^2(Q)^2 + \omega^2 \gamma^2(Q)},
$$

(2)

where $\chi(Q) \equiv \chi'(Q) \equiv \chi'(Q, \omega = 0)$ is the real part of the zero frequency susceptibility. The solution of Eq. 1 can be represented by two poles with complex frequencies:

$$
\omega = \pm \left[ \omega_0^2 - \left( \frac{\gamma^2}{4} \right) \right]^{\frac{1}{2}} = \pm \omega_1 - \frac{i \gamma}{2}.
$$

(3)

If $\omega_0^2 \geq \gamma^2/4$, $\omega_1$ is real and the frequency of the pole. The solutions (response) correspond to damped oscillations in time. If $\omega_0^2 \leq \gamma^2/4$, $\omega_1$ is imaginary and the system is overdamped. We may introduce a third frequency, $\omega_{\text{max}}$, defined as the frequency at the peak in $\chi''(\omega)$. This can be shown to be

$$
\omega_{\text{max}} = \frac{1}{6} \sqrt{12 \omega_0^2 - 6 \gamma^2 - 6 \sqrt{\gamma^4 - 4 \gamma^2 \omega_0^2 + 16 \omega_0^4}}.
$$

(4)

Using the DHO function (Eqn. 2) to analyse all of the data allows a consistent model to be applied to the underdamped and overdamped regimes. This is useful when comparing excitations from undoped and doped compositions.

### III. RESULTS

#### A. RIXS spectra of La$_{2-x}$Sr$_x$CuO$_4$

Fig. 1 (c) shows example spectra from each composition at $Q = (0.2, 0)$. The low-energy magnetic spectrum of the parent ($x = 0$) compound (bottom), is dominated by resolution-limited spin-wave excitations. The magnetic excitations in the doped $x = 0.12$ (middle) and $x = 0.16$ (top) compositions are considerably broader as noted in previous studies. The $dd$ excitations occur in the energy range 1–3 eV.

Figs. 3 and 4 show examples of our RIXS data. Spectra such as those in 3 and 4 are collected together into intensity maps plotted as a function of $Q$ and energy in Fig. 2. Thus Fig. 2 gives an overall picture of the excitations observed in the present study. The strongest feature in Fig. 2(a) is the magnon which disperses to an energy $\sim 355 \pm 34$ meV along $(h, 0)$ in agreement with previous studies. The magnetic excitations are much broader
FIG. 4. Examples of fitted RIXS spectra from LCO and LSCO $x = 0.12$ (performed at I21 at DLS) and $x = 0.16$ (performed at ID32 at the ESRF). Showing data in the high $Q$ regime from high symmetry directions $(h, 0), (h, h)$. The data have intensity $I_{\text{RIXS}}$ indicating that they are normalised to an integration over the range of the $dd$ excitations, $g$. The total fit to the data is indicated in red, the DHO magnetic excitations in pink, elastic peak in green, phonon excitations in yellow and dark blue, multi-magnons in purple and background in light blue.

in energy for doped compositions as shown in Figs. 2(c)-(f). Phonons can be seen in the $\text{La}_2\text{CuO}_4$ spectra below 100 meV, for example in Fig. 2(c) and also visible in the map plots in Fig. 2. In Fig. 2(c), for $x = 0.12$ again a phonon branch can be seen below 100 meV along $(h, 0)$ in addition to CDW order near $h = 0.23$. Similar behaviour is seen in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8 + \delta$.

In addition to the high-symmetry direction measurements show in Figs. 2(c)-(f), a full quadrant of the Brillouin zone was examined by mapping $(h, k)$ in the $x = 0.16$ and 0.12 compounds. Approximately 90 spectra were collected at the ID32 beamline, distributed throughout the zone with spacing 0.05 (r.l.u.). The RIXS intensity is plotted as a function of $(h, k)$ for several energy slices and for $x = 0.12$ in Fig. 5 where areas of high-intensity correspond to the spin-excitation intensity. These measurements were performed with lower resolution ($\Delta E \approx 50$ meV). The plots are smoothed by averaging neighbouring points within $|\Delta Q| = 0.05$ (r.l.u.). At low energies, the maximum in the RIXS intensity appears at low $Q$ and is approximately symmetrically distributed around $\Gamma$. As the energy increases, peaks develop along $(h, 0)$ and $(0, k)$ and move to larger $h$ and $k$. It is interesting to note that quite similar behaviour is observed in $\text{La}_2\text{CuO}_4$, where $(h, k)$ maps measured with INS show a peak in the intensity at $(\frac{1}{2}, 0)$ for energies above about 320 meV. The maps show that for doped LSCO the magnetic spectral weight persists to higher energies near $(\frac{1}{2}, 0)$ than in other parts of the Brillouin zone. This observation is consistent with previous work.

B. DHO fitting

Figs. 3 and 4 show fits of the damped harmonic oscillator (DHO) model (Sec. II D) together with phonon peaks and background to the data. The 35 meV resolution of the instrument allows the phonons and elastic peaks to be separated from the DHO response. As can be seen from the figures, the DHO model generally works well. The measured spectra are shown in black with the total fitted function indicated in red with constituent functions below. At low $Q$ the excitations are hard to model due to their proximity to the elastic peak and the comparatively low energy and intensity of the spin wave poles. The grey region in Figs. 7 and 8 indicates this low $Q$ regime where the excitations are underdamped and have multiple solutions to Eqn. 2. The parameters $\omega_0$ and
γ/2 extracted from DHO fits are plotted for Q = (h, 0) and (h, h) in Fig. 7 for each compound. Hole doping in the doped compounds, it is comparable to ω₀. The damping is anisotropic in wavevector space, that is γ/2 is larger along (h, h) than along (h, 0). Our data also reveals that the anisotropy of the damping does not reflect the antiferromagnetic Brillouin zone as γ/2 peaks at approximately (0.2, 0.2) [rather than (1/4, 1/4)] along (h, h). This effect can be seen both for x = 0.12 and x = 0.16 Fig 7(d,f).

We also fit the lower resolution (ΔE ≃ 50 meV) spectra from the grid in (h, k). The results of fitting this data to the DHO model are summarised in Fig. 6. The damping γ/2 is again seen to be largest in the region near (0.2,0.2) for both doped compositions. The over-damped region where ω₀² < γ²/4 and ω₁ is imaginary, is indicated in Fig. 6(d,i) as ω₁ = 0.

C. Estimate of the absolute wavevector-dependent susceptibility

Fitting our RIXS data to the DHO response function in Eqn. 2 allows the wavevector-dependent susceptibility χ′(Q) to be estimated, where

\[ χ′(Q) = χ′(Q, ω = 0) = \frac{1}{π} \int_{-∞}^{∞} χ′′(Q, ω) dω. \]  

In this section, we estimate χ′(Q) in the superconductors we have investigated by using the parent antiferromagnet La₂CuO₄ as a reference. The analysis discussed so far has relied on normalisation to g, an integration over the region of the dd excitations, to take account of angle-dependent effects on the RIXS intensity. This does not affect the determination of excitation energies or damping coefficients. However, this procedure does not account for the difference in absorption between photons scattered from the magnetic excitations, which are close to the resonance, and the dd-excitations which are significantly away from the Cu absorption peak (width of about 0.4 eV) and are therefore less likely to be absorbed. In order to correct for these effects, we use the measured spin wave RIXS intensity of the parent compound as a reference. INS measurements show that the magnetic excitations of La₂CuO₄ are fairly well-described by linear spin wave theory (SWT) with some corrections²⁰ near (1/4, 1/4). Thus the underlying S(Q, ω) is known in this case.

Ament et al.²⁰ point out that under certain theoretical approximations, the absolute RIXS cross-section can be split into a prefactor f(ε, ϵ′, k, k′) multiplied by a dynamic structure factor S(Q, ω), where the polarisations of the initial and final photons are ε and ϵ′. We note that the exact circumstances when the RIXS response is proportional to S(Q, ω) is still an active subject of investigation²¹, however, we will use this approximation in our analysis. Here we propose a simple estimate to remove the effects of f(ε, ϵ′, k, k′) from S(Q, ω) for doped LSCO. We assume f(ε, ϵ′, k, k′) is the same for doped and undoped compounds. For each (k, k′) we first normalise (divide) the raw RIXS spectra by g to yield IRIXS (see Sec. IID) and find χ′ RIXS by fitting to the DHO model. We then multiply χ′ RIXS for LSCO by the spin-wave response of LCO determined from INS²¹ divided by the measured RIXS response of LCO, to estimate the dynamic susceptibility of the doped superconductor in absolute units:

\[ \langle χ′ LSCO(Q) \rangle = χ′ RIXS(Q) × \frac{ϕ LCO(SWT)}{ϕ RIXS(Q)}. \]  

ϕ LCO(SWT) is the energy integrated spin-wave pole weight, determined from a fit of linear SWT to INS data and ϕ RIXS is the integrated pole weight of fitted RIXS data, details of this are given in appendix A. In practice, we fit the LCO spectra and then use Eqs. A4 and A7 to evaluate ϕ LCO(SWT) and ϕ RIXS. Eqn. 6 assumes that the factors f and g are the same in doped and undoped compositions and therefore cancel in the normalisation procedure. We have verified that this is approximately the case in our samples.

Figs. 7 and 8(a) and (b) show the parameters γ(Q), ω₀(Q) and χ′(Q) extracted from fits of Eq. 2 as a function of Q along (h, 0) and (h, h) for the three compounds. For LCO, χ′(Q, ω) is a sum of two contributions with the form of Eqn. 2 to account for the single-magnon and
multi-magnon excitations. The resulting $\chi'_{\text{RIXS}}(Q)$ due to the single-magnon pole is shown in Fig. 6 (a) and (b) with a cubic polynomial fit indicated with a solid blue line. The susceptibilities $\chi'_{\text{RIXS}}(Q)$ in Fig. 6 (a) and (b) contain the effects of the $f$ factor and self absorption mentioned above. In Fig. 8 (c) and (d) we correct for these effects and estimate the absolute $\chi'(Q)$ using Eqs. 6, A4 and A7 together with the cubic polynomial fit of $\chi'_{\text{RIXS}}(Q)$ to La$_2$CuO$_4$ in Fig. 8 (a,b).

By definition, the corrected susceptibility for the parent compound La$_2$CuO$_4$ becomes that of the SWT model described in Appendix A and verified by INS. For all three compositions investigated, $\chi'(Q)$ increases as we move along (h, h) towards $(\frac{1}{4}, \frac{1}{2})$, where INS finds the strongest spin fluctuations. The magnitude of $\chi'(Q)$ is larger in the doped compositions $x = 0.12, 0.16$ than in the parent, this effect is also present when the data is normalised via the $dd$ excitations so does not seem to be an artefact arising from the spin wave normalisation. It arises because spectral weight is moved to lower energy which gives a larger contribution to $\chi'(Q)$ (see Eqn. 5).

The resulting modelled excitations are shown in Fig. 9 where $\chi''(Q, \omega)$ is calculated from Eqn. 2 with the fitted parameters $[\omega_0(Q), \gamma(Q), \chi'(Q)]$ shown in Figs. 7 and 8.

IV. DISCUSSION

A. Theoretical Models

Our investigation of the magnetic excitations in cuprates is motivated by spin-fluctuation mediated theories of high temperature superconductivity and to gain a fundamental understanding of metallic transition metal oxides. The Hubbard model (in its one or three band variants) is generally considered to be a good starting point. Calculations based on the Hubbard model show that the wavevector-dependent pairing interaction is proportional to $\chi'(Q)$. Numerical studies of the two-dimensional Hubbard model, applied to cuprates, qualitatively reproduce the slowly-evolving high-energy magnetic excitations which are observed by INS and RIXS experiments, but calculations are restricted to relatively small lattices. Other approaches based on renormalised itinerant quasiparticles with various types of approximation provide a basis for a phenomenological understanding of the physical properties and allow finer structure in wavevector and energy to predicted. In general, we expect the magnetic excitations and $\chi''(Q, \omega)$ to be different around (0, 0) and $(\frac{1}{2}, \frac{1}{2})$ and the dispersion of the excitations not to be symmetric around $(\frac{1}{4}, \frac{1}{2})$.

B. Wavevector dependence of the response

The high-energy magnetic excitations in the parent compound La$_2$CuO$_4$ are anisotropic in two ways. Firstly the single magnon energy varies between points on the
antiferromagnetic Brillouin zone boundary with \((\frac{1}{2}, 0)\) having a higher energy than \((\frac{1}{4}, \frac{1}{4})\). Secondly, the single magnon excitation is strongly and anomalously damped at the \((\frac{1}{2}, 0)\) position. This variation in the magnon energy can be understood in terms of an expansion of the single band Hubbard model\(^{19,45}\) which gives rise to second nearest neighbour and cyclic exchange interactions. While the anisotropy of the damping in La\(_2\)CuO\(_4\) may be understood in terms of the unbinding of magnons into spinons\(^{20,46}\).

Our data show how the anisotropies of the parent compound persist into the doped compositions and are qualitatively consistent with previous studies\(^{11,32}\). However, the higher energy resolution of the present study \((\Delta E \approx 35 \text{ meV})\) as compared to \(\Delta E \gtrsim 100 \text{ meV}\) in previous work\(^{11,13,26}\) allows us to separate the magnetic excitations from lower energy features. In Fig. 7, we see that the frequency of the undamped mode \(\omega_0(Q)\) extracted from the DHO model shows similar dispersions along \((h, 0)\) and \((h, h)\) in the doped \(x = 0.12\) and \(x = 0.16\) compositions as in the parent \(x = 0\). At \(Q = (\frac{1}{2}, 0)\), \(\hbar \omega_0\) increases with doping from \(355 \pm 34 \text{ meV} \) \((x = 0)\) to \(416 \pm 41 \text{ meV} \) \((x = 0.16)\), while at \(Q = (\frac{1}{2}, \frac{1}{2})\) it increases from \(297 \pm 28 \text{ meV}\) to \(317 \pm 34 \text{ meV}\).

A new result from this work is the extent of the variation of \(\gamma(Q)\) and \(\omega_0(Q)\) across the Brillouin zone in doped LSCO. Significantly, the damping is seen to increase in the underdoped compound, \(x=0.12\) and again in the optimally-doped material, \(x=0.16\). From the damping maps shown in Fig. 6 (b) and (g) it can be seen that the enhanced damping is most prominent close to the \((h, h)\) direction. It is notable that the maxima in \(\gamma(Q)\) and \(\omega_0(Q)\) along \((h, h)\) are actually near \((0.2, 0.2)\) rather than at \((\frac{1}{2}, \frac{1}{2})\). Our \((h, k)\) maps of the fitted parameters in Fig. 8 show that the \(\gamma(Q)\) actually shows a local maximum around this point. These features appear to be qualitatively present in theoretical calculations based on itinerant quasiparticle such as those in Refs. 13 and 44 and presumably arise from (nesting) features in the underlying quasiparticle band structure. The general damping anisotropy between \((h, 0)\) and \((h, h)\) for the doped compositions has also been described by theories based on determinantal quantum Monte Carlo (DQMC)\(^{31}\).

The normalisation procedure described in Sec. IIIC

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**FIG. 7.** Summary of fit parameters to a damped harmonic oscillator response model as a function of wave-vector \(Q\) along high symmetry directions \((h, 0)\) and \((h, h)\). \(\omega_0\) is indicated with blue circles and the damping coefficient \(\gamma/2\) is shown as red squares. Errors are from fitting considering the standard error in the raw data. Solid lines are a cubic polynomial fit to the data. Data in panel (a), (b) and (c) all contain data measured at I21 and panel (c) contains additional data measured at ID32. The dashed grey line marks the AF Brillouin zone boundary.

**FIG. 8.** Wavevector-dependent susceptibilities \(\chi'(Q)\) in \(\text{La}_{2-x}\text{Sr}_x\text{CuO}_4\) determined from RIXS spectra. Fits of a damped harmonic oscillator function to \(I_{\text{RIXS}}\) yield estimates of \(\chi'_{\text{RIXS}}(Q)\) shown in (a) and (b) which include self-absorption and other orientation-dependent effects. (c) and (d) show estimates of the absolute \(\chi'(Q)\). These estimates are obtained by normalising the data from doped compositions by the antiferromagnetic parent compound as described in the text. Cubic polynomial fits to the data are shown as solid lines and the dashed line shows the SWT model. The dashed grey line indicates the Brillouin zone boundary. Data on all compounds were collected at I21 and additional data on the \(x = 0.16\) compound were measured at ID32.
that spin fluctuation in the theoretical calculations based on the Hubbard model INS measurements change interactions and is qualitatively consistent with is expected because of the residual antiferromagnetic ex-

M the antiferromagnetic Brillouin zone centred on h,h χ which shifts spectral weight to lower energy. Also of cause of the smaller χψ(Q) is about 4 times larger at (1 1 0) than at (1 0 0). This arises because of the smaller ωψ(Q) at (1 1 0) (see Figs. 4 and 7) which shifts spectral weight to lower energy. Also of interest is the fact that χψ(Q) increases monotonically along (h, h) from Γ to M. The increase is consistent with the fact that the magnetic response is strongest in the antiferromagnetic Brillouin zone centred on M. This is expected because of the residual antiferromagnetic-exchange interactions and is qualitatively consistent with INS measurements. It should be noted that theoretical calculations based on the Hubbard model show that spin fluctuation in the M zone contributes most to pairing in spin-fluctuation mediated theories of HTC.

C. Comparison to high-energy INS

RIXS and INS provide complementary views of the collective spin excitations in the cuprates. However, INS measurements of the high-energy magnetic excitations are difficult because the background increases when high incident energies are used. Nevertheless, some data does exist for La$_{2-x}$Sr$_x$CuO$_4$. An early study on La$_{1.86}$Sr$_{0.14}$CuO$_4$ revealed magnetic excitations up to 260 meV. In particular, excitations were observed at Q = (1 2 0) which is equivalent to the Q = (1 0 0) position investigated here with RIXS. Our RIXS normalisation procedure (Sec. III C) allows us to estimate χψ(Q) = 1.52 ± 0.34µ$_B^2$ eV$^{-1}$ f.u.$^{-1}$ in LSCO x = 0.16 at (1 2 0) based on an integration of the spectrum up to about 800 meV. Integrating the INS data in Ref. 15 up to 260 meV we obtain χψ(Q) ≈ 0.5 µ$_B^2$ eV$^{-1}$ f.u.$^{-1}$. Thus, if it were possible to perform neutron scattering experiments over a wider energy range the integration of INS data may produce a comparable value for χψ(Q) at q = (1 2 0). The approximate agreement is satisfying, however further work is required to develop the comparison of the two probes of collective magnetic excitations.

V. SUMMARY AND CONCLUSIONS

We have made high-resolution RIXS measurements of the collective magnetic excitations for three compositions of the superconducting cuprate system La$_{2-x}$Sr$_x$CuO$_4$. Specifically, we have mapped out the excitations throughout the 2-D (h, k) Brillouin zone to the extent that is possible at the Cu-L edge. In addition, we have attempted to determine the wavevector-dependent susceptibility of the doped compositions La$_{2-x}$Sr$_x$CuO$_4$ (x = 0.12, 0.16) by normalising data to the parent compound. This procedure allows comparison with INS measurements. We find that the evolution of the intensity of high-energy (hω ≥ 200 meV) excitations measured by RIXS and INS is consistent.

The high-energy spin fluctuations in La$_{2-x}$Sr$_x$CuO$_4$ are fairly well-described by a damped harmonic oscillator model. The DHO damping parameter increases with doping and is largest along the (h, h) line although it is not peaked at the high symmetry point (1 1 1). While the pole frequency is peaked at (1 0 0) for doped and undoped compositions, for the doped compositions, the wavevector-dependent susceptibility χψ(Q) is much larger at (1 1 0) than at (1 0 0). Both of these positions are on the antiferromagnetic zone boundary of the parent compound. The wavevector-dependent susceptibility increases rapidly along the (h, h) line towards the antiferromagnetic wavevector of the parent compound (1 1 1). Thus the strongest magnetic excitations and those predicted to favour superconductive pairing occur towards the (1 1 1) position. Our quantitative determination of the wavevector-dependent susceptibility will be useful in testing magnetic mediated theories of high-temperature superconductivity.

Appendix A: Linear spin-wave theory calculations

The magnetic excitations can be modelled in LCO with classical linear spin-wave theory. We consider the case of a S = 1/2 square lattice antiferromagnet with nearest-
and next-nearest exchange interactions. The susceptibility transverse to the ordered moment \( \chi''(Q, \omega) \) due to one-magnon creation is given by:

\[
\chi''(Q, \omega) = Z_d(Q) \frac{\pi}{2} g^2 \mu_B^2 S \left( \frac{A_Q - B_Q}{A_Q + B_Q} \right)^{1/2} \delta[\omega + \omega_0(Q)],
\]

where

\[
h\omega_0(Q) = 2 Ze \sqrt{A_Q^2 - B_Q^2},
\]

and

\[
\chi'_\perp(Q) = Z_d(Q) \frac{g^2 \mu_B^2 S}{A_Q + B_Q}.
\]

The amplitude factors \( A_Q \) and \( B_Q \) are given by \( A_Q = J - J_c/2 - (J' - J''/4)(1 - v_k v_k)/2 \) and \( B_Q = (J - J_c/2)(v_k + v_k)/2 \), where \( v_k = \cos(2\pi x) \) and \( x \to k \). \( Z_d \) and \( Z_e \) are renormalisation constants which take account of quantum fluctuations in the AF ground state.

Headings et al.\(^1\) have made INS measurements of the spin waves in La\(_2\)CuO\(_4\) and fitted the model described by Eqs. (A1)-(A3). They find \( J = 143, \ J' = 2.9 \) and \( J_c = 58 \) meV, assuming \( Z_e = 1.18 \). The wavevector dependence of \( Z_d(Q) \) is also determined. In order to compare the INS and RIXS measurements, we assume that RIXS is equally sensitive to the three components of the susceptibility and compute the average susceptibility \( \chi = \frac{1}{3} (\chi_{xx} + \chi_{yy} + \chi_{zz}) = \frac{2}{3} \chi_{\perp} \). The energy integrated intensity of the spin wave pole \( \phi^{\text{LO}}_{\text{SWT}}(Q) \) is then:

\[
\phi^{\text{LO}}_{\text{SWT}}(Q) = \frac{\pi}{3} \chi''(Q, \omega) d\omega = \frac{\pi}{3} \chi'(Q) \omega_0(Q). \quad \text{(A4)}
\]

We derive a comparable measure of the energy integrated spin-wave pole measured with RIXS by rewriting Eqn. (2) for LCO (in the limit \( \omega_0 \geq \gamma/2 \)) as,

\[
\chi''(Q, \omega) = \frac{\chi'(Q)}{2\omega_1(Q)} \left( \frac{\gamma^2(Q)}{4} + \omega_1^2(Q) \right) \times \left\{ \frac{\gamma(Q)^2/2}{\gamma^2(Q)/4 + [\omega - \omega_1(Q)]^2} - \frac{\gamma(Q)^2/2}{\gamma^2(Q)/4 + [\omega + \omega_1(Q)]^2} \right\}.
\]

Integrating over the positive energy pole, we obtain the measured pole intensity from the fitted parameters \( \omega(Q), \gamma(Q) \) and \( \chi'(Q) \):

\[
\phi^{\text{LO}}_{\text{RIXS}}(Q) = \frac{\pi \chi'(Q) \omega_0^2(Q)}{\sqrt{4\omega_0^2(Q) - \gamma^2(Q)}}. \quad \text{(A7)}
\]

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