

# Evidence for spin-fluctuation-mediated superconductivity in electron-doped cuprates

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# **Evidence for spin-fluctuation-mediated superconductivity in electron-doped cuprates**

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Supplementary Information

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## Comparison of zero-field resistivity in different films

Two films, S1 ( $0.116 \leq x \leq 0.159$ ) and S2 ( $0.159 \leq x \leq 0.188$ ), were patterned into microbridges and their in-plane resistivity measured using a standard four-point ac lock-in method. Panel a) of Fig. S1 shows a comparison of the  $\rho(T)$  curves from film S1 with previous data reported in ref. (1) at similar doping levels. We find that the overall resistivity values in the new films are approximately 1/3 of those reported in ref. (1) and their residual resistivity ratios (RRR) are also higher. There are two probable reasons for these observations. Firstly, in ref. (1), the film thickness was estimated from the fabrication conditions, likely leading to deviations from the real value. In the present study, a step profiler was used to measure the thickness, so in principle the as-determined thickness should be closer to the actual value. (We note that a similar multiplicative factor appears in the comparison of the Hall coefficients shown in Extended Data Fig. 6.) These variations in the absolute magnitude of  $\rho(T)$  do not affect the analysis in ref. (1) since all the reported data were obtained on a single film with the same nominal thickness. Secondly, continual improvement in the quality of the combinatorial films has led to larger RRR and slightly higher  $T_c$  values, possibly reflecting improved crystallinity or a more homogeneous oxygen content. Crucially, when the data are scaled such that their  $\rho(T)$  curves have the same slope and then adjusted vertically to account for the differences in their residual resistivities (Fig. S1b), the curves overlap.

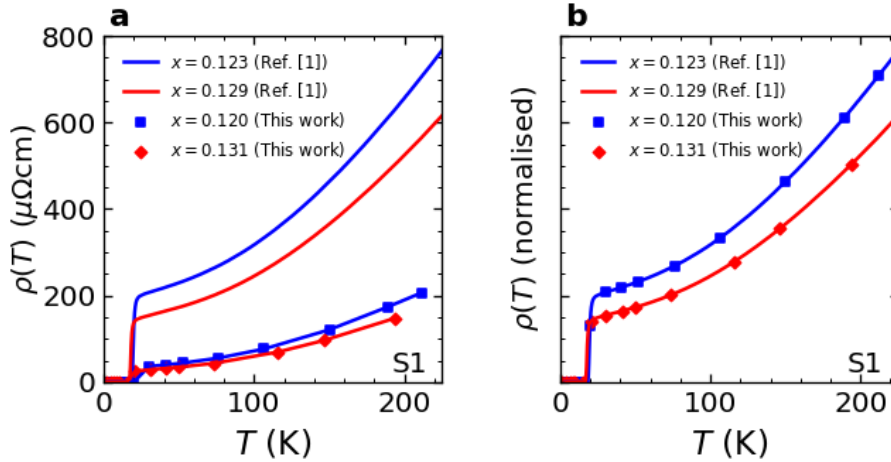


**Figure S1: Zero-field resistivity of overdoped LCCO films.** (a) As-measured  $\rho(0, T)$  data on S1 (symbols) with data for comparable  $x$  values from Ref. (1). (b) Closer comparison of the two data sets having scaled the slopes of each curve in Ref. (1) and shifted them vertically to account for differences in their residual resistivities.

## Link between $T$ -linear scattering rate and $T_c$ in metals

To highlight the link between the strength of the  $T$ -linear scattering rate and  $T_c$  in ordinary metals, we focus here on the work of Allen<sup>2</sup> in which a near-equivalence of the coupling strengths deduced from  $T_c$  ( $\lambda_{ph}$ ) and from the coefficient of the  $T$ -linear resistivity  $A_1$  ( $\lambda_{tr}$ ) was

demonstrated for a wide array of elements and alloys. For most elements considered in Allen's treatise,  $T_c$  was derived directly from the McMillan formula:

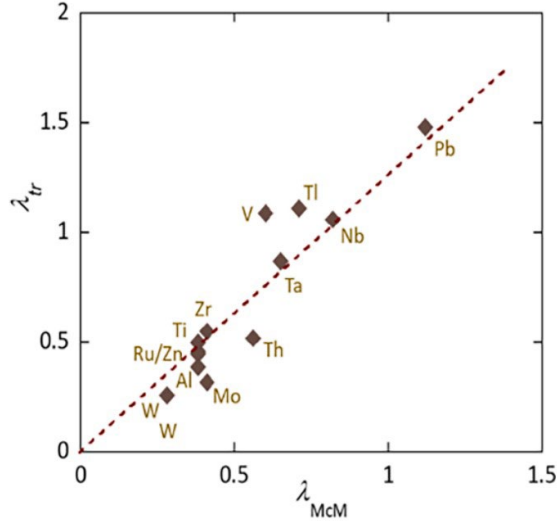


**Figure S2:  $T$ -linear resistivity and  $T_c$  in conventional metals.** Comparison of  $\lambda_{tr}$  – the coupling strength deduced from the  $T$ -linear resistivity and  $\lambda_{McM}$  – the e-ph coupling strength deduced from the value of  $T_c$  in a subset of elemental superconductors using the McMillan formula (taken from Ref. (3)).

$$T_c = \frac{\theta_D}{1.45} \exp \frac{-(1.04(1+\lambda_{McM}))}{(\lambda_{McM} - \mu^*(1+0.62\lambda_{McM}))} \quad (1)$$

assuming the same value for the pseudopotential  $\mu^*$  consistent with tunneling and theory. Hence, a determination of  $\lambda_{McM}$  required only knowledge of  $T_c$  and the Debye temperature  $\theta_D$ . For  $\lambda_{tr}$ , Allen used density functional theory to determine the FS and density of states for the individual elements and then used the Drude equation to deduce the strength of the scattering rate  $\Gamma_{tr}$  from  $A_1$ . Once this was obtained, Allen determined  $\lambda_{tr}$  directly from the expression  $\hbar\Gamma_{tr} = 2\pi\lambda_{tr}k_B T$ . Extended Data Fig. 2 shows a comparison of  $\lambda_{tr}$  and  $\lambda_{McM}$  for some elemental superconductors. Despite some scatter around the dotted line in SI Fig. 2, the overall trend is clear and indicates a robust correlation between  $\lambda_{tr}$  and  $\lambda_{McM}$ , and through this, between  $T_c$  and  $A_1$  for almost one decade is variation in  $\lambda_{tr}$  ( $\lambda_{McM}$ ). Hence, while there are undoubtedly complicating circumstances, the electron-phonon coupling strength  $\lambda_{e-ph}$  does indeed appear to play the decisive role in determining  $T_c$  and  $A_1$ .

In the simulations in Figure 4 of the main article, the form of  $\beta(\varphi)$  in Eq. (1) of the main article for the spin fluctuation scenario had the form ( $= \sin^{12}[2\varphi \pm (\pi/4 - HS)]$ ). Obviously, the higher the exponent  $n$  in a  $\sin^n(2\varphi)$  parameterisation, the more sharply peaked the maxima in  $\varphi$ . The choice of  $n = 12$  for the spin-fluctuation scenario was made for two reasons: (i) it produces well-defined hotspots at the AFM zone boundary, with clear minima between adjacent hotspots, and (ii) its form is such that the minima in  $\tau^{-1}(\varphi)$  at the nodes are higher than those the antinodes. This second aspect is crucial in the subsequent discussion of the Hall response, since it is precisely this disparity between the nodes and anti-nodes that enables the  $T$ - (and  $x$ -) dependence of the Hall

coefficient to be captured with the same ansatz (once current vertex corrections are also taken into account – see below).

Nevertheless, any function that satisfies these two criteria would have the same effect on the MR. To demonstrate that it is the overall form of the scattering rate that is important, Fig. S3 compares simulations for 20 K and 50 K (the extremes of our fitting range) performed using the  $\sin^{12}$  function (as presented in the main article) and a Gaussian function of similar width and height. In both cases the overall magnitude and form of the MR is the same, with slight differences between the two forms of the scattering rate responsible for minor discrepancies. Note that changing the width of the anisotropic component (i.e. increasing the width of the Gaussian, or reducing the power of the  $\sin$  function) will have an impact on the resulting simulations, as it is the anisotropy of the scattering rate that controls the magnitude and linearity of the MR itself.

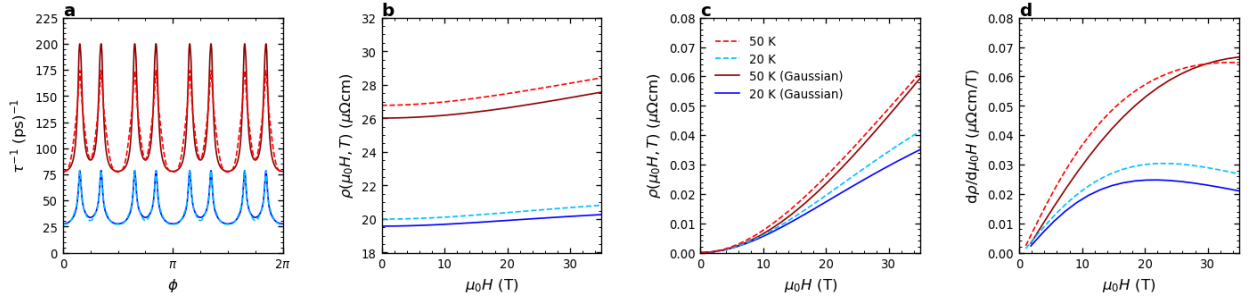


**Figure S3: Sinusoidal vs. Gaussian functional form of the scattering rate.** (a) The scattering rate at 20 K (blue) and 50 K (red) using the  $(= \sin^{12}[2\phi \pm (\pi/4 - HS)])$  form from the manuscript (dashed lines) and a Gaussian form (solid lines) with a similar height and width. Note that the 50 K scattering rate has been offset by 50  $\text{ps}^{-1}$  for clarity. (b) The corresponding MR simulation in absolute values, (c) normalised to  $\rho(0)$ , and (d) its derivative, clearly showing comparable behaviour between the two. Small differences between the two can be attributed to slight differences in the scattering rate.

As mentioned in the main article and based on our analysis of the Hall effect in overdoped LCCO (presented in the proceeding section), we conclude that only a scenario consistent with strong scattering off commensurate AFM spin fluctuations can satisfactorily and self-consistently account for all the observed magnetotransport behaviour and in the remainder of this section, we only consider this case. In order to model the  $x$ -dependence of the MR and  $\rho(0, T)$ , we assume that it is the  $T$ -linear component of  $\Gamma_T$  that changes with  $x$ , its variation defined by the dimensionless parameter  $g(x)$  whose absolute values are listed in Extended Data Table 3. Comparison of the raw  $\Delta\rho/\rho(0)$  data and simulations for other doping levels studied on film S1 at  $T = 40$  K are shown in Fig. S4. We find that the evolution of both the  $H$ -dependence and magnitude of  $\Delta\rho/\rho(0)$  with  $x$  can be well described simply by varying  $g(x)$ . Due to the extent of  $\Gamma_T$  in momentum space, scaling  $g(x)$  additionally affects the zero-field resistivity. The small discrepancy between the simulated and measured absolute values of the MR shown in Fig. 5b) of the main article could be removed by imposing fewer constraints on the scattering rate, though for simplicity, we have chosen not to do this.

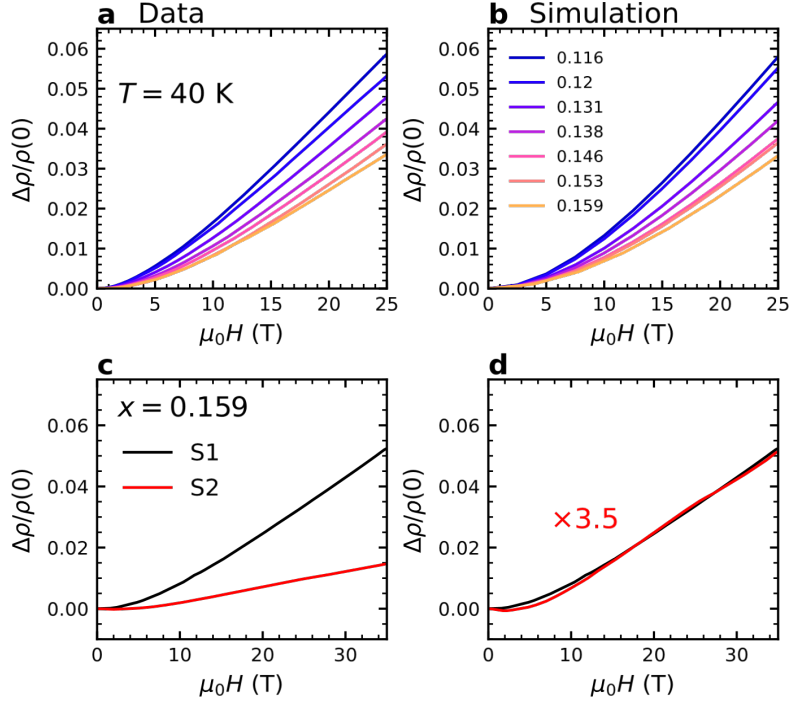


**Figure S4: Magnetoresistance and simulations for selected dopings from film S1.** **a)** The experimentally obtained  $\Delta\rho/\rho(0)$  for 7 doping values on S1 at  $T = 40$  K. **b)** The simulations for the same doping levels as in (a) where only  $g(x)$  has been varied to capture the  $x$ -dependence. **c)** The unscaled and **d)** scaled MR of S2 at  $x=0.159$ . The origin of the reduced MR in S2 is most likely the negative MR component that is observable at low field strengths. This low-field negative MR contribution is seen in all channels in S2. As a result, we only use the zero-field resistivity, and more specifically, the evolution of its  $T$ -linear coefficient  $A_1$ , to deduce the value of  $g(x)$  beyond  $x = 0.159$ .

As  $x$  increases, it is expected that the hotspots broaden or diminish in size, in tandem with  $g(x)$ , as one might expect upon approaching the FL regime. Indeed, we find that increasing the width of the hotspot by a factor of two or decreasing the size of the hotspot by a factor of 5 reduces the resistivity by  $1 \mu\Omega\text{cm}$ . This behaviour may explain the larger increase of  $g(x)$  in our simulations below  $x = 0.14$  and at lower  $T$ : the sharpening and strengthening of hotspots would contribute to the increase in the resistivity in parallel with the anisotropic  $T$ -linear term. Note that the strength of the hotspot can only be quantified experimentally if breakdown in the form of MR saturation is observed. Thus, for the sake of transparency and simplicity, we keep the height of  $\tau_{\text{HS}}^{-1}$  constant at all  $x$  and  $T$ . The origin and nature of  $\Gamma_T$  will be discussed further and in more detail below. Finally, as mentioned in the main article, the MR of each voltage pair in S2 was found to contain a negative component, visible in Fig. S4 for the raw and scaled data, respectively. This made it difficult to model the MR for  $x > 0.159$  without introducing an additional scaling factor. For this reason,  $g(x)$  for  $x > 0.159$  was determined solely from the evolution of the  $T$ -linear coefficient of  $\rho(0, T)$  (i.e. the extrapolated  $T = 0$  intercept of Extended Data Fig. 1c), having first scaled the value of  $g(x)$  for  $x = 0.159$  for S2 to that obtained in S1.

## Effect of CVCs on the in-plane magnetoresistance

In Fig. S5, we show that the incorporation of modified velocities (from spin fluctuation scattering) into the SCTIF has a negligible effect on the modeling of the MR. (Note that the same modified velocities – expressed in Eqs. (11) and (12) – were included in the SCTIF Boltzmann calculations in the same way as for the Zener-Jones estimation.) From panels a) and b), it is clear that the effect on the MR is small; this is again evident in panel c) where the 20 K curves from both simulation are overlaid. Panel d) also demonstrates only a modest difference in  $\rho(0, T)$ . The effect of CVCs on the MR is small since (a) the MR is not influenced by the circulation of  $\ell$  around the FS (in contrast to the Hall conductivity), and (b) the MR is determined largely by those sections of FS with the longest  $\ell$ , i.e. away from the hotspots. At the hotspots themselves,  $\ell$  is extremely short and thus, those regions provide a negligible contribution to the total (magneto)-conductivity, even if their presence does have a strong influence on the  $H$ -dependence of the MR (through impeded cyclotron motion). Hence, the inclusion of CVCs does not affect the resistivity or the low-field MR in a significant way.

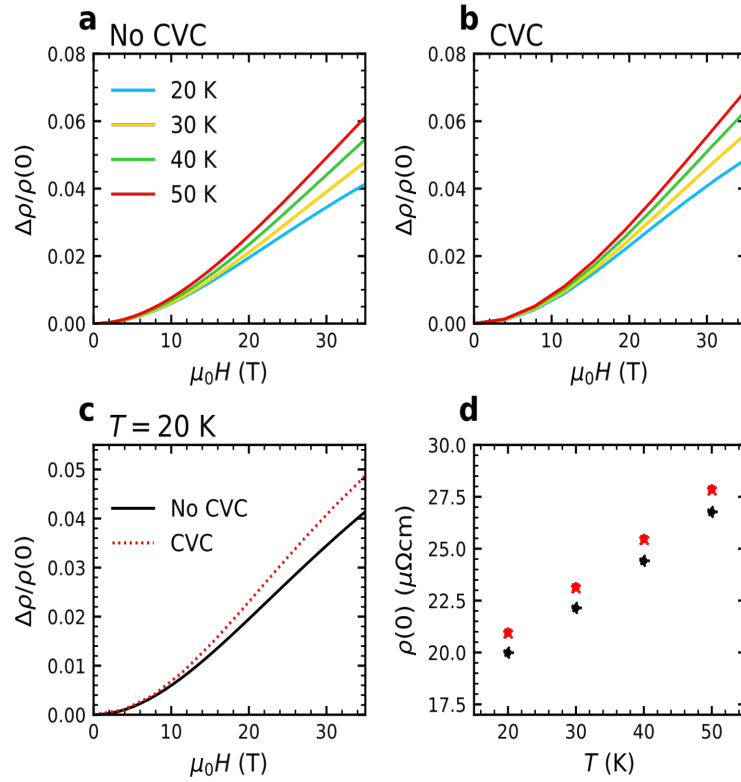


**Figure S5: Effect of current vertex corrections on the MR and zero-field resistivity.** (a) The MR for  $x = 0.159$  without using CVCs, as in the main article. (b) The same with CVCs. (c) The MR at 40 K without (black) and with (red) CVCs. (d) The zero-field resistivity without (black) and with (red) CVCs. Incorporating CVCs into the simulations of the MR has almost no effect on the magnitude and form of the MR, nor does it significantly impact the zero-field resistivity.

## Inclusion of inelastic processes within Boltzmann

Within the Boltzmann approach, quasiparticles travel on their cyclotron orbits without changing their energy and without decaying into other particles. In other words, only the momentum (not the energy) of the quasiparticle decays. Nevertheless, Boltzmann theory, specifically within the relaxation time approximation (RTA), has been used to model the resistivity of metals effectively for over a century. In reality, Boltzmann transport within the RTA cannot differentiate between elastic and inelastic scattering. Boltzmann cares only about the relaxation of ‘velocity’ (through the vector  $\ell$ ) – no matter if energy is exchanged in the process - just like it cannot distinguish between phonons, magnons, impurities or quasi-particle lifetimes from imaginary self-energies. In some sense, this is a disadvantage as it provides no direct information (we infer spin here because the hotspots can emerge from the interaction of the conduction electrons with finite- $Q$  spin waves and because the inclusion of CVCs associated with these spin waves enables the  $T$ -dependence of the Hall response to be also captured, and we infer inelastic because  $T$ -activated phenomena naturally involve energetics). At the same time, it is also an advantage in the sense that Boltzmann is agnostic, because otherwise we would have even more fitting parameters and procedures. How momentum exactly relaxes in dc transport and if this relaxation involves a Planckian bound is an important open question and, no matter if we treat copper or cuprates, remains an active field of research.

In his seminal work treating spin fluctuation scattering near a QCP in the presence of disorder, Rosch applied a Boltzmann approach incorporating transition rates associated with both elastic impurity scattering and inelastic scattering from spin fluctuations<sup>4</sup>. Rosch argued that this approach works provided that the spin fluctuations are assumed to stay in equilibrium, an approximation that is valid if the spin fluctuations lose their momentum effectively by Umklapp or impurity scattering. These are the same conditions we are assuming here.

## Supplementary Information References

1. Yuan, J. et al., *Nature* **602**, 431 (2022).
2. Allen, P. B. *Handbook of Superconductivity* (Academic Press, San Diego, USA, 2000), pp. 478–489.
3. Hussey, N. E. *Physica C* **614**, 1354362 (2023).
4. Rosch, A. *Phys. Rev. Lett.* **82**, 4280 (1999).