Extreme Suppression of Antiferromagnetic Order and Critical Scaling in a Two-Dimensional Random Quantum Magnet

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Doping quantum magnets with various impurities can give rise to unusual quantum states and quantum phase transitions. A recent example is Sr$_2$CuTeO$_4$, a square-lattice Néel antiferromagnet with superexchange between first-neighbor $S = 1/2$ Cu spins mediated by plaquette centered Te ions. Substituting Te by W, the affected impurity plaquettes have predominantly second-neighbor interactions, thus causing local magnetic frustration. Here we report a study of Sr$_2$CuTe$_{1-x}$W$_x$O$_4$ using neutron diffraction and μSR techniques, showing that the Néel order vanishes already at $x \approx 0.03$. We explain this extreme order suppression using a two-dimensional Heisenberg spin model, demonstrating that a W-type impurity induces a non-collinear deformation of the order parameter that decays with distance as $1/r^2$ at temperature $T = 0$. Thus, there is a logarithmic singularity and loss of order for any $x > 0$. Order for small $x > 0$ and $T > 0$ in the material is induced by weak inter-plane couplings. In the non-magnetic phase, the μSR relaxation rate exhibits quantum critical scaling with a large dynamic exponent, $z \approx 3$, consistent with a random-singlet state.

In the field of quantum magnetism, studies of impurities and disorder (randomness) have traditionally complemented investigations of translationally invariant systems. The success of this approach is epitomized by spin chains with random couplings, which were discovered experimentally \[1\] \[2\] and led to the development of the strong-disorder renormalization method \[3\] and the concept of the random singlet (RS) state \[4\] \[5\]. There is a long-standing quest to identify two-dimensional (2D) analogues \[6\] \[8\] of the RS state, and promising model systems have been identified that may harbor this type of non-magnetic randomness-induced state \[9\] \[16\] with universal quantum-critical scaling properties \[14\] \[17\] \[18\]. Possible experimental signatures of RS physics have been reported in frustrated 2D quantum magnets such as YbMgGaO$_4$ \[18\] and $\alpha$-Ru$_{1-x}$Ir$_x$Cl$_3$ \[19\], but the properties of these systems are affected by anisotropic Dzyaloshinskii-Moriya interactions and spin vacancies, respectively, in addition to the random Heisenberg couplings. Beyond the intrinsic interest in the 2D RS state as an exotic randomness-induced spin liquid, this state should also be a useful benchmark for experiments on potential uniform spin liquids \[20\] \[21\], where it is often difficult \[10\] \[18\] \[22\] \[23\] to distinguish between impurity physics and theoretically predicted properties of clean systems.

An interesting emergent candidate material for unperturbed 2D RS physics is Sr$_2$CuTe$_{1-x}$W$_x$O$_4$. At $x = 0$, this layered system can be well described by the prototypical 2D $S = 1/2$ antiferromagnetic (AFM) Heisenberg model with

![](https://example.com/figure1.png)

Figure 1. 2D Heisenberg couplings $J_1 S_i \cdot S_j$ in Sr$_2$CuTe$_{1-x}$W$_x$O$_4$. The small black circles represent the $S = 1/2$ carrying Cu ions, while red and blue circles correspond to Te and W ions, respectively. The dominant couplings mediated by Te in (a) and W in (b) are first-neighbor $J_1$ (solid red lines) and second-neighbor $J_2$ (solid blue lines), with $J_1 \approx J_2 \approx 8$ meV \[29\] \[32\]. The couplings $J_1'$ and $J_2'$ indicated by the thin dashed lines are roughly 10% of the dominant couplings. The first-neighbor coupling $J_1'$ on links between Te and W ions, the gray dashed line in (c), is about 4% of $J_1$ \[32\].
predominantly first-neighbor interactions $J_1$, which are generated through a superexchange mechanism via Te ions located at the centers of the plaquettes of $2 \times 2$ Cu ions \cite{24,25}, as illustrated in Fig. 1(a). At $x = 1$, W-mediated superexchange instead leads to second-neighbor (diagonal) couplings between the spins on the affected plaquettes, Fig. 1(b), with strength $J_2$ close to the value of the Te-mediated $J_1$ \cite{26-28}. The entire range $x \in [0,1]$ can be realized, and an intriguing magnetically disordered state exists within a window $[x_{c1}, x_{c2}]$ \cite{29-31}. The coupling constants have been calculated also in the mixed case \cite{29,32}; see Fig. 1(c). The ability to tune the combined degrees of frustration and disorder by $x$ offers unique opportunities to systematically study a randomness-induced state, possibly the 2D RS state, arising from a well-defined Hamiltonian that can in principle be solved with state-of-the-art numerical quantum many-body methods \cite{12,15,16}.

Not only is the putative RS state itself interesting, but the continuous variation of $x$ also allows for studies of the quantum phase transition out of the $J_1$-dominated Néel AFM state into the magnetically disordered state at $x = x_{c1}$, and out of this state into the $J_2$-dominated columnar AFM state for $x = x_{c2}$. Experiments on $\text{Sr}_2\text{CuTe}_{1-x}\text{W}_x\text{O}_6$ reported to date indicate $x_{c1} \approx 0.1$ and $x_{c2} \approx 0.6$ \cite{29,30}, but these quantum phase transitions remain unexplored.

We here report $\mu$SR and neutron diffraction experiments, demonstrating that the Néel order in $\text{Sr}_2\text{CuTe}_{1-x}\text{W}_x\text{O}_6$ vanishes at smaller $x$ than previously believed, at $x_{c1} \approx 0.03$. To explain this dramatic order suppression, we study a classical Heisenberg model with the estimated couplings in $\text{Sr}_2\text{CuTe}_{1-x}\text{W}_x\text{O}_6$ and random locations of W and Te ions. We conclude that the Néel order at temperature $T = 0$ in a 2D plane is destroyed even at infinitesimal $x$, due to a previously unknown logarithmic singularity caused by a non-colinear deformation of the bulk colinear order parameter. Order at $x > 0$ and $T > 0$ is stabilized by weak inter-layer couplings, which are expected in $\text{Sr}_2\text{CuTe}_{1-x}\text{W}_x\text{O}_6$ and explain the quantum phase transition at $x_{c1} \approx 0.03$. Starting from $x = 1$ the columnar AFM state is much more stable, which also can be explained by the classical model. In the non-magnetic phase, the neutron diffraction measurements reveal strong short-range Néel correlations and the $\mu$SR relaxation rate exhibits quantum-critical scaling with dynamic exponent $z > 2$, both consistent with the 2D RS scenario \cite{14}.

Polycrystalline samples of $\text{Sr}_2\text{CuTe}_{1-x}\text{W}_x\text{O}_6$ were synthesized from stoichiometric mixtures of SrO, CuO, TeO$_2$, and WO$_3$ powders by the solid-state reaction reported previously \cite{24,25,26,28}. The $\mu$SR experiments were performed at the S1 ARTEMIS spectrometer, J-PARC, with the mini cryostat. The neutron-diffraction experiments were carried out on Bamboo ($\lambda = 2.358$ Å) and Xingzhi ($\lambda = 2.7302$ Å) triple-axis spectrometers, and at the PKU High-Intensity Powder Neutron Diffractometer ($\lambda = 2.3$ Å) at China Advanced Research Reactor (CARR), and the Kunpeng triple-axis spectrometer ($\lambda = 2.7302$ Å) at Key Laboratory of Neutron Physics and Institute of Nuclear Physics and Chemistry, China.

Figure 2 shows our neutron diffraction results. For the $x = 0$ and $x = 1$ samples, Figs. 2(a) and 2(e), resolution-limited magnetic peaks are observed, which is consistent with previous reports of long-range order \cite{24,25}. Substituting W in the $x = 1$ sample by Te up to 30% does not alter the position of the magnetic peaks, Figs. 2(f) and 2(h), and they remain resolution-limited, suggesting the presence of long-range columnar AFM order in all these samples. In contrast, while the very lightly W doped sample with $x = 0.02$, Fig. 2(b), is still long-range Néel ordered with sharp resolution limited peaks, the peaks for $x = 0.1$ and 0.2 in Figs. 2(c) and 2(d) are already much broader than the instrumental resolution, indicating the loss of long-range order somewhere between $x = 0.02$ and 0.1. We also note that the magnetic correlation lengths of the $x = 0$ and 0.02 samples exceed 180 Å (about 35 lattice spacings) according to the instrument resolution, while that of the $x = 0.1$ sample is about 40 Å.

Figures 3(a) and 3(b) show zero-field $\mu$SR spectra of $\text{Sr}_2\text{CuTe}_{1-x}\text{W}_x\text{O}_6$ at $x = 0$ and 0.05, respectively. The data...
Asymmetry $A_0 / A_0 (30\text{K})$ \begin{table}[h]
\begin{tabular}{|c|c|c|c|}
\hline
$\lambda (\text{µs}^{-1})$ & 0.10 & 0.20 & 0.25 \\
\hline

\end{tabular}
\end{table}

Figure 3. Zero-field $\mu$SR spectra of $\text{Sr}_2\text{CuTe}_{1-x}\text{W}_x\text{O}_6$ samples with $x = 0$ in (a) and $x = 0.05$ in (b). Results for several temperatures are shown, with the highest and lowest indicated, and in between these $T/\text{K} = 12.5, 18.7, 21.2, 22.1, 25.5, 27.5$ in (a) and $10.6, 18.9, 25.5, 27.5$ in (b). The curves are fits to Eq. (1). The temperature dependent asymmetry $A_0$, normalized by the value at $T = 30\text{K}$, is shown for samples with $x = 0, 0.05$, and $0.1$ in (c) and for $x = 0.7$ and $1$ in (d). The relaxation rates are shown in (e) for the samples with phase transitions and in (f) for those with only short-range correlations. In (f), the fitted lines correspond to critical scaling, $\lambda \propto T^{-\gamma}$, with $\gamma = 0.23 \pm 0.03$ for $x = 0.05$ and $0.39 \pm 0.03$ for $x = 0.1$.

In contrast, in the $x = 0.05$ and $0.1$ samples, Fig. 3(c), $A_0$ only decreases slowly below a characteristic temperature $T^*$ (see also Supplemental Information). This behavior reflects gradual changes of the local fields as a result of the onset of short-range magnetic correlations but no ordering, which for $x = 0.1$ is consistent with the neutron results in Fig. 2(c).

The relaxation rate $\lambda$ also exhibits different behaviors in the samples with phase transitions, Fig. 3(e), and short-range correlations, Fig. 3(f). The power-law behaviors for $x = 0.05$ and $0.1$ reflect quantum-critical scaling in what is likely the RS phase. As explained in Supplemental Information, standard scaling arguments $[34,35]$ in combination with a constraint imposed by the $1/r^2$ form of the spin correlations in the RS phase $[14]$ can be used to derive the form $\lambda \propto T^{-\gamma}$ with $\gamma = 1 - 2/\varepsilon$. The values of $\gamma$ extracted from the fits in Fig. 3(f) correspond to $\varepsilon = 2.6 \pm 0.3$ for $x = 0.05$ and $\varepsilon = 3.3 \pm 0.3$ for $x = 0.1$. These values of the dynamic exponent conform with the expectations in the RS phase, where $\varepsilon$ equals 2 at the Néel–RS transition and grows upon moving.
inside the RS phase \cite{14}. At $x = 0.7$ in Fig. 3(e), the small upturn below $T_c$ may indicate close proximity to the RS phase.

Combining the $\mu$SR and neutron-diffraction results, the magnetic phase diagram of Sr$_2$CuTe$_{1-x}$W$_x$O$_6$ is shown in Fig. 4(a). We conclude that the effects of doping on the $x = 0$ and $x = 1$ AFM system are drastically different, as reflected in the sizes of the Néel and columnar phases. The columnar order is robust even for large Te substitution, which is indicative of only minor effects of magnetic frustration and remaining large connected ordered regions. The mean order parameter may then be gradually reduced in a way similar to diluted systems with continuous phase transitions \cite{36}. In contrast, introducing W in the $x = 0$ sample rapidly destroys the Néel order somewhere between $x = 0.02$ and 0.05. Based on the trends in the neutron and $\mu$SR data sets, we estimate the quantum phase transition at $x_{c1} = 0.03 \pm 0.01$. Short-range correlations with Néel structure still remain even at $x = 0.2$ based on our neutron-diffraction experiments and likely remain throughout what we have argued is the RS phase.

While the size of the columnar AFM phase agrees with other studies \cite{29,32}, the size of the Néel phase in Fig. 4(a) is smaller by at least 50% than the previous estimates. The fact that the Néel phase is smaller than the columnar phase can be understood from a simple argument based on the dominant coupling constants $J_1$ and $J_2$ illustrated in Fig. 1. Introducing a single Te impurity in the $J_2$ coupled columnar system, we simply lose the $J_2$ couplings in the affected plaquette and there is only a weak frustration effect from the much smaller $J_{1}'$ and $J_{2}'$ couplings. However, when introducing a W impurity in the $J_1$ dominated Néel state, the two new $J_2$ bonds are completely frustrated. Thus, it is not surprising that the Néel phase at finite W doping fraction is smaller than the columnar phase at finite Te doping, but to quantitatively understand the extremely narrow Néel phase requires further insights.

Ideally, we would like to carry out an unbiased calculation with the full quantum mechanical Heisenberg Hamiltonian. Even though progress has been made on some 2D quantum magnets with density-matrix renormalization group (DMRG) \cite{37} and tensor-product \cite{38} methods, including for Heisenberg systems with random couplings \cite{10}, in practice calculations are still challenging and it would be hard to extract a reliable phase diagram. However, we have found that already the classical Heisenberg model can explain the extreme fragility of the Néel state to W plaquette impurities, and also give an overall reasonable phase diagram.

The Heisenberg model with uniform exchange $J_i S_i \cdot S_j$ on all first neighbors $(i,j)$ is of course long-range ordered at $T = 0$. According to the Mermin-Wagner theorem \cite{39}, in two dimensions the order is destroyed by thermal fluctuations at any $T > 0$. In weakly coupled planes, the critical temperature takes the form $T_c \propto J / \ln(J/J_c)$, where $J_c$ is the coupling between spins in adjacent planes \cite{40,41}. These general results apply to quantum as well as classical spins, and a quantum spin system with AFM order or a long correlation length behaves in many respects as a “renormalized” classical system \cite{42}. One can therefore expect the initial effects of doping the $x = 0$ and $x = 1$ system to be captured correctly by a classical model, up to factors close to 1.

In the notation of Fig. 1 we set the 2D couplings to $J_1 = J_2 = 1$, $J_1' = J_2' = 0.1$, and $J_{2}'' = 0$, with $|S_z| = 1$. We have also checked that small variations in the parameters do not significantly affect the results. For coupled planes we take $J_{1} = 10^{-3}$ and $10^{-3}$. We used standard Monte Carlo methods for frustrated Heisenberg models \cite{35,44}, with Binder cumulant techniques \cite{45} for extracting the critical temperature at fixed $x$. Results for $0 < x < 1$ were averaged over several hundred realizations of the random W and Te plaquettes on systems with up to $64 \times 64 \times 32$ spins. The resulting infinite-size extrapolated phase boundaries are shown in Fig. 4(b). When comparing with the experiments, it should be noted that $T = 25$ K corresponds roughly to 0.3 in units of $J_1$ and that $T_c$ in uniform coupled $S = 1/2$ planes with $J_{1}$ of order $10^{-2}$ is lower by about 50% than our classical result at $x = 0$ \cite{41}. We expect quantum fluctuations to shrink the size of the two ordered phases also in the $x$ direction.

As seen in Fig. 4(b), upon changing $J_{1}$ from $10^{-2}$ to $10^{-3}$, $T_c$ at $x = 0$ is only slightly reduced, as expected on account of the logarithmic form discussed above \cite{40,41}. For $x > 0$ the phase boundary drops more rapidly to zero for the smaller $J_{1}$, and the size of the Néel phase is substantially smaller. A very narrow Néel phase with high sensitivity of the $T = 0$ transition point to $J_{1}$ are not natural features within a simple picture of conventional local impurity suppression of the order \cite{36}. It is therefore useful to investigate the deformation of the Néel order around a single impurity plaquette at $T = 0$, which we have done by minimizing the energy with a combination of simulated annealing and energy conserving spin moves.

The deviation $\Delta m$ of the local ordered moment from the bulk value $m = 1$ is graphed in Fig. 5 versus the distance...
r from the impurity. The magnitude of the deformation decays as 1/r^2, which causes a logarithmic divergence when integrated over r (but the total energy cost of the deformation stays constant, with the energy density decaying as 1/r^4). This single-impurity response implies that any fraction x > 0 of impurities destroys the long-range order, as demonstrated explicitly in Supplemental Information. Such an effect was previously believed to be possible only in frustrated systems with non-collinear bulk order [46], but our results show unambiguously that the plaquette impurity has this profound unexpected effect on the collinear Néel order. The 1/r^2 deformation persists also if there is no frustration at all in the bulk system (J'_1 = 0 instead of J'_2 = 0.1 used in Fig. 5), as shown in the Supplementary Information.

In the model system of weakly coupled planes, corresponding to the phase diagram in Fig. 2(b), the 3D couplings stabilize the Néel order for a range of x > 0 depending on J'_1/J, but we have not studied the functional form of x_c1 versus J'_1. In an S = 1/2 system such as Sr_2CuTe_1-xW_xO_6, quantum fluctuations should further suppress the order and reduce x_c1, and we expect the same type of logarithmic singularity as in the classical case when J'_1/J → 0, on account of the renormalized classical picture of the quantum Néel state [42].

The singular distortion effect of the W impurities in the Néel state are not captured by previous treatments such as density functional theory, which predicts the critical fraction x_c1 ≈ 0.1 [29]. Though we expect classical and quantum spin systems to have qualitatively similar initial impurity responses, the disordered classical and quantum low-temperature phases should differ qualitatively. In the classical case, in general a spin glass is expected, as recently discussed in the context of bond-random J_1-J_2 Ising [47] and triangular-lattice Heisenberg antiferromagnets [46]. In the presence of strong quantum fluctuations, as in Sr_2CuTe_1-xW_xO_6, there is mounting evidence from model studies that the spin glass in general is supplanted by an RS state [13,14,16,46]. Extensive studies of the RS state realized with a specific S = 1/2 model uncovered quantum criticality with a large dynamic exponent and dominant Néel-type spin correlations decaying universally with distance as 1/r^2 at T = 0 [14,17]. This form of the correlations was also recently detected in a frustrated random-bond system with DMRG calculations [16].

We conclude that the presence of significant Néel correlations in Sr_2CuTe_1-xW_xO_6, revealed by our neutron results in Fig. 2 at x = 0.1 and 0.2, well past the loss of long-range order at x ≈ 0.03, is expected within the RS scenario. Previous results at x = 0.5 also show some remaining Néel features [32]. Moreover, the µSR relaxation rate at x = 0.05 and 0.1 (in Fig. 3f) exhibits quantum-critical scaling with dynamic exponent z > 2, which is also expected in the RS state [44,17]. Thus, overall our experimental results lend support to the RS scenario for x ∈ [0.05, 0.20]. It was also previously shown that the temperature dependence of the susceptibility [30] agrees with the RS predictions [14] for x ∈ [0.2, 0.5]. Most likely, the entire range x ∈ [0.03, 0.6] where no order is detected realizes the RS phase with a varying z > 2.

It would be interesting to further test the proposed RS scaling forms experimentally in Sr_2CuTe_1-xW_xO_6. Detailed inelastic neutron scattering studies would be very useful, but our attempts to grow large single-crystals have so far not been successful. With polycrystalline samples, NMR experiments may be able to further elucidate the nature of the lightly W-doped distorted Néel state and the RS state.

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Supplementary Information

Extreme Suppression of Antiferromagnetic Order by Local Frustration Effects in a Two-Dimensional Quantum Magnet

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Here we provide additional experimental, theoretical, and Monte Carlo simulation results supporting the conclusions of the main paper. In Sec. 1 [1] we explain how the cross-over temperature $T^*$ was determined from the neutron diffraction data. In Sec. 2 [2] we discuss the $\mu$SR relaxation rate $\lambda$, deriving its expected quantum-critical form using standard scaling arguments and the properties of the RS phase previously established in models [14,17]. In Sec. 3 we present additional Monte Carlo results demonstrating that the long-range order in the classical 2D Heisenberg model at $T = 0$ vanishes for any concentration $x$ of the W-type frustrated plaquette impurities, due to a logarithmic singularity in the $x \to 0$ limit.

1. THE CROSS-OVER TEMPERATURE $T^*$

For the samples showing no phase transition into an ordered phase, in Fig. 4 we have indicated a temperature $T^*$ where both the $\mu$SR and neutron data show the onset of significant short-range correlations. It should be noted that, strictly speaking, $T^*$ can not be defined unambiguously or uniquely as it merely signifies a sharp cross-over. Therefore, $T^*$ determined from the neutron-diffraction measurements is not necessarily exactly equal to that from the $\mu$SR data, since these two techniques measure the system in different ways and with very different energy resolution. We here show that both experiments nevertheless produce compatible results for $T^*$.

Figures SI[a] and SI[b] show the temperature dependence of the magnetic peak intensity measured with neutron diffraction at wave-vector $q = (0.5, 0.5, 0)$ (corresponding to Néel AFM order) for the $x = 0.1$ and $0.2$ samples, respectively. $T^*$ is determined to be the temperature where a signal is detected above the high-$T$ background, which is $T^* \approx 25$ and $T^* \approx 6$ K, respectively, for $x = 0.1$ and $x = 0.2$, with rather large error bars of $2\pm3$ K due to the weak signal. Comparing with the $\mu$SR result for $x = 0.1$ in Fig. 4, the results agree well. We do not have $\mu$SR results for $x = 0.2$.

2. CRITICAL SCALING OF THE RELAXATION RATE

As discussed in the main paper, the $x = 0.05$ and $0.1$ samples exhibit quantum-critical scaling in the $\mu$SR relaxation rate and are candidates for the RS state at low temperatures. According to QMC simulations of a “designer model” realizing the RS phase in a 2D quantum magnet [14,17], this state is critical with rather large dynamic exponent, $z \geq 2$, and with dominant Néel type spin correlations decaying with distance $r$ as $r^{-z}$. This correlation function formally implies that the exponent $\eta$ in the standard form [34] of the quantum-critical
correlation function for a system in \( d \) space dimensions,

\[
C(r) \propto r^{-(d+z-2+\eta)}, \tag{S1}
\]
depends on \( z \) through the relationship \( \eta = 2-z \). Thus, in the RS state this exponent is negative, which is normally not possible in uniform systems but is not uncommon in disordered systems. The exponent \( \eta \) appears also in some dynamical scaling forms, e.g., the NMR relaxation rate \( 1/T_1 \) scales as \( T^n \) at the O(3) quantum-critical point in uniform antiferromagnets, where \( z = 1 \) \cite{55}. One can expect that the \( \mu\)SR relaxation rate \( \lambda \), which like \( 1/T_1 \) depends on local low-energy spin fluctuations, should scale in the same way. However, since the dynamic exponent \( z \neq 1 \) in the RS state, the \( T^n \) form has to be modified as follows: The correlation length in a quantum-critical system scales as \( \xi \propto T^{-1/z} \), and we can therefore formally express the temperature as \( T \propto \xi^{-z} \). For \( z = 1 \), we can write \( \lambda \propto T^n \propto \xi^{-\eta} \), and the generalization to \( z \neq 1 \) is obtained by inserting the correct \( T \)-dependent expression for the correlation length. Thus, \( \lambda \propto \xi^{-\eta} \propto T^{n/z} \). Using the form \( \eta = 2-z \) in the RS state, we expect \( \lambda \propto T^{-\gamma} \), where we have defined the positive exponent \( \gamma = 1-2/z \), with \( z \geq 2 \), which was extracted from the data fits in Fig. 3(f).

The asymptotic scaling form of \( \lambda(T) \) can also be derived in a more transparent way: First, consider the well known NMR spin-lattice relaxation rate \( 1/T_1 \), which for a spin-isotropic system is given by \cite{48}

\[
\frac{1}{T_1} = \frac{\gamma^2}{2} \sum_q A^2(q) S(q, \omega_N), \tag{S2}
\]

where \( \gamma \) is the gyromagnetic ratio, \( A_q \) is the Fourier transform of the hyperfine constants describing the coupling between the nuclear and electronic spins, and \( \omega_N \) is the field-dependent nuclear resonance frequency. The hyperfine coupling is short-ranged in space, and if the nucleus considered is in the ion hosting the localized electronic spins (e.g., Cu NMR in the material considered here), it is often sufficient to consider purely local on-site interactions \( A_q \), so that the momentum sum in Eq. (S2) reduces to \( A^2_0 S_0(\omega_N) \), where \( S_0(\omega) \) is the on-site (single-spin) dynamic structure factor.

Typically, the resonance frequency is much lower than other energy scales in the system, and the zero-frequency limit can be considered (unless there are significant spin diffusion contributions, which can cause low-frequency divergencies). Thus, with these simplifications, which are often completely valid, the relaxation rate is proportional to \( S_0(\omega \rightarrow 0) \) (with prefactors that are known or can be measured). Since \( \mu\)SR also is a probe of low-frequency local spin fluctuations, we expect the same form;

\[
\lambda \propto S_0(\omega \rightarrow 0). \tag{S3}
\]

The local dynamic spin structure factor \( S_0(\omega) \) (and also its \( q \) dependent variant) can be calculated in various analytical approximative schemes or numerically; for example, it was calculated in the case of the 1D RS state in Ref. \cite{49}. However, the low-frequency limit is often challenging, especially in QMC calculations, where the corresponding imaginary-time dependent spin correlation function \( G_0(\tau) \) has to be calculated and analytically continued to real frequency. To circumvent the latter step, Randera et al. suggested a very useful approximation \cite{50}, which was expressed in a slightly different form in Ref. \cite{49}. Neglecting unimportant factors, the approximation amounts to \( S_0(\omega \rightarrow 0) \propto G_0(\tau = \beta/2)/T \), which implies that the relaxation rate \( \lambda(S) \) is approximated as

\[
\lambda \propto \frac{1}{T} G_0(\tau = \beta/2), \tag{S4}
\]

where \( \beta = 1/T \). Here we will use this form, which is expected in general to become better with decreasing \( T \), to derive the critical scaling behavior of \( \lambda \) in the RS phase.

As already mentioned above, a quantum-critical spatial correlation function is conventionally written as Eq. (S1), where \( d = 2 \) in our case. The on-site correlation in imaginary time is modified by the dynamic exponent \( \eta \)

\[
G_0(\tau) \propto \tau^{-(d+z-2+\eta)/z}, \tag{S5}
\]

reflecting that space (and imaginary) time distances are related as \( \tau \propto r^z \), which is used to obtain Eq. (S5) from Eq. (S1). Thus, in the RS state with the staggered spatial spin correlation function \( C(r) \propto r^{-z} \), the time correlations take the form

Figure S1. Temperature dependence of the magnetic peak intensity at \( q = (0.5, 0.5, 0) \) in the sample with \( x = 0.1 \) in (a) and \( x = 0.2 \) in (b). The curves are guides to the eye and the cross-over temperature is defined as the point where the signal above background becomes significant, which implies errors of up to 3 K in these cases.
$G_0(r) \propto r^{-2/2}$. Using this form in Eq. (S4) immediately gives the scaling form $\lambda \propto L^{-(1-2/z)}$, in agreement with the result presented earlier. The fact that we observe this kind of scaling with $z > 2$, Fig. [S3f], with $z$ also increasing upon moving further away from the Néel phase [14], constitutes strong support for an RS phase in Sr$_2$CuTe$_{1-x}$W$_x$O$_6$.

### 3. 2D HEISENBERG MODEL

In Fig. [5] in the main text we demonstrated an impurity induced deformation of the sublattice magnetization that decays with the distance $r$ from the impurity as $1/r^2$. This decay implies that the total response of a single impurity diverges logarithmically with increasing system size. We here provide additional results demonstrating that the order parameter indeed vanishes for any concentration $x > 0$ of the impurities.

In the main paper, the Monte Carlo simulations were carried out with parameters approximating those estimated [32] for Sr$_2$CuTe$_{1-x}$W$_x$O$_6$. The bulk parameters for $x = 0$, illustrated in Fig. [1a], were $J_1 = 1$ and $J_2 = 0.1$. Even with the small frustrating $J_2$ terms, the $T = 0$ order parameter is the fully collinear Néel state, and we do not expect that the frustration is in any way required to obtain the $r^{-2}$ decay of the deformation. To explicitly demonstrate that the classical Heisenberg model with only the first-neighbor couplings $J_1$ also has the same impurity response as in Fig. [5] here in Fig. [S2] we show simulation results for $J_2 = 0$. These results confirm that the $r^{-2}$ form emerges as the system size increases. For the following results we go back to $J_2 = 0.1$, and we expect the same kinds of behaviors also for $J_2 = 0$.

In Fig. [S3a] we show results for the disorder-averaged $T = 0$ Néel order parameter $m$ versus the concentration of impurities. Increasing the system size consistently leads to a smaller value of $m$. In Fig. [S3b] we show results versus the inverse system size for several low impurity concentrations. Here we can observe that $m$ always decreases with increasing $L$. Given the logarithmic singularity suggested by the single-impurity response, the most natural scenario is that $m$ vanishes in the thermodynamic limit for all $x > 0$, but it is difficult to demonstrate that reliably using results such as those in Fig. [S3] because of the logarithmic-type singularity that makes extrapolations difficult.

A better way to investigate the presence or absence of order for small $x$, introduced in Ref. [51] is to consider a system with a single impurity to have concentration $x = 1/L^2$, and to compute the initial slope $R = dm/dx$ of the order parameter vs $x$ based on this value;

$$R_1(L) = L^2[1 - m_1(L)],$$

where $m_1$ is the value of $m$ computed with the single impurity (averaged over the entire system). Then, if indeed $m = 0$ for $L \rightarrow \infty$ at $x = 0^+$, the slope $R_L(L)$ will diverge. In order to take into account possible subtle interaction effects, we here additionally use a modified approach with $L$ randomly placed impurities in the $L^2$ system, for which the concentration is $x = 1/L$ and the slope is

$$R_L(L) = L[1 - m_1(L)],$$

where $m_L(L)$ is the impurity-averaged order parameter for $L$ impurities in the lattice with $L^2$ spins. In Fig. [S4a] we show $m_1(L)$ and $m_L(L)$ versus $1/L$. In the former, we can see clearly the expected approach to the fully saturated bulk order parameter $m = 1$ when $L$ increases. For $m_L(L)$ we also have to asymptotically approach the same limit, and this appears plausible though the convergence is slower, as expected, because of the higher concentration $x$ for a given system size. In Fig. [S4a] we graph the initial
slopes defined in Eqs. \((S6)\) and \((S7)\). Both quantities diverge logarithmically, confirming that the impurity response in the \(x \to 0\) limit has a logarithmic singularity. Any other interpretation than \(m(x) = 0\) for all \(x > 0\) is then unlikely, as indicated also by the results in Fig.\( S4\) for small but finite impurity concentrations.

The Néel order suppression for any \(x > 0\) is also supported by the strong sensitivity of \(T_c(x)\) to the 3D coupling \(J_{\perp}\) in Fig. \(4\) (b), which suggests that the transition into the ordered phase at \(x > 0\) and \(T > 0\) is due to the inter-layer effect. It would be interesting to also study the deformation induced by a single-impurity in the 3D coupled-layer system, but we have not yet done so. We should expect the \(1/r^2\) decay to be cut off at some distance depending on \(J_{\perp}\) (diverging as \(J_{\perp}/J_1 \to 0\)) and, therefore, the slopes defined in Eqs. \((S6)\) and \((S7)\) to be finite for any \(J_{\perp} > 0\). Related issues were recently discussed by Dey et al. in the context of a host system (the Heisenberg model on the triangular lattice) with coplanar AFM order \(46\).

Figure S4. (a) Néel order parameter vs inverse system size in systems with a single impurity (blue symbols) and with \(L\) impurities (red symbols), graphed versus the inverse of the system size \(L\). (b) Slope graphed on a log-linear plot of the magnetization curve at \(x = 0\) based on the size-dependent definitions, Eqs. \((S6)\) and \((S7)\), with the data in (a). The lines are fits corresponding to the logarithmically divergent forms \(R_n(L) \sim a_n + b_n \log(L)\) with both definitions (with systems containing \(n = 1\) and \(n = L\) impurities).