Low temperature ballistic spin transport in the \( S = 1/2 \) antiferromagnetic Heisenberg chain compound \( \text{SrCuO}_2 \)

H Maeter\(^1\), A A Zvyagin\(^1,2\), H Luetkens\(^3\), G Pascua\(^3\), Z Shermadini\(^3\), R Saint-Martin\(^4\), A Revcolevschi\(^4\), C Hess\(^5\), B Büchner\(^5\) and H-H Klauss\(^1\)

\(^1\) Institute for Solid State Physics, TU Dresden, D-01069 Dresden, Germany
\(^2\) Institute for Low Temperature Physics and Engineering of the NAS of Ukraine, Kharkov, 61103, Ukraine
\(^3\) Laboratory for Muon-Spin Spectroscopy, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland
\(^4\) Laboratoire de Physico-Chimie de L'Etat Solide, ICMMO, UMR 8182, Université Paris-Sud, F-91405 Orsay, France
\(^5\) IFW-Dresden, Institute for Solid State Research, PO Box 270116, D-01171 Dresden, Germany

E-mail: h.klauss@physik.tu-dresden.de

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Abstract

We report zero and longitudinal magnetic field muon spin relaxation (\(\mu\)SR) measurements of the spin \( S = 1/2 \) antiferromagnetic Heisenberg chain material \( \text{SrCuO}_2 \). We find that in a weak applied magnetic field \( B_0 \) the spin–lattice relaxation rate \( \lambda \) follows a power law \( \lambda \propto B_0^{-n} \) with \( n = 0.9(3) \). This result is temperature independent for \( 5 \text{ K} \leq T \leq 300 \text{ K} \). Within conformal field theory and using the Müller ansatz we conclude ballistic spin transport in \( \text{SrCuO}_2 \).

The copper oxide based low-dimensional electronic systems allow a detailed study of many cooperative quantum phenomena and concepts, such as Mott insulators, spin–charge separation, quantum phase transitions, and unconventional superconductivity [1–5]. Quasi 1D arrangements of corner-sharing \( \text{CuO}_2 \) squares are model compounds for the \( S = 1/2 \) antiferromagnetic Heisenberg chain (AFHC). In these systems, charge degrees of freedom are quenched at low energies by strong Coulomb interactions. Spin degrees of freedom are governed by the Heisenberg Hamiltonian \( H = J \sum_i S_i S_{i+1} \), where \( i \) indexes the spins along the chain and the exchange constant \( J \) controls the interaction strength between neighboring spins. A model compound for the isotropic \( S = 1/2 \) AFHC is \( \text{SrCuO}_2 \). It is regarded as an almost ideal 1D system with \( J \approx 2100 \text{ K} \) between neighboring spins of magnetic \( \text{Cu}^{2+} \) ions [6]. In relation to \( J \) the residual interchain interaction that causes magnetic order at temperatures below \( \approx 2 \text{ K} \) [7] is very small.

The ground state of the \( S = 1/2 \) AFHC is a singlet (\( S = 0 \)) state with a continuum of \( S = 1/2 \) excitations—spinons. Recently, using numerical solutions of the Bethe ansatz equations, it was shown that the ground state dynamic correlation functions are determined mostly by the 2-spinon continuum [8–11]. While its thermodynamic properties have been studied thoroughly, both theoretically and experimentally, the dynamic properties, such as the spin transport, have been much less studied: for a strongly interacting system with a non-equidistant (due to interactions) spectrum of eigenstates, it is an arduous task. As a result, theoretical studies are often controversially discussed, and even diffusive spin transport has been predicted for some temperature and field regimes [12–18].

Recent heat transport experiments revealed an unexpectedly large magnetic contribution to the total heat conductivity of \( S = 1/2 \) AFHC materials such as \( \text{SrCuO}_2 \), attributed to the magnetic excitations of the spin chains at low temperatures [19–21]. It is still unclear how this contribution can be understood microscopically. A deeper understanding of the dynamic properties of the prototype Hamiltonian for the \( S = 1/2 \) AFHC, the Heisenberg Hamiltonian \( H = J \sum_i S_i S_{i+1} \),
is essential for progress in this field. Its most important property is integrability, i.e., the existence of infinitely many local constants of motion. In general, integrability implies ballistic (spin) transport [22]. In particular, ballistic spin transport is predicted for finite magnetic fields and the ground state [17, 18]. However, an experimental proof for ballistic spin transport in the $S = 1/2$ AFHC is still lacking. Several experimental studies reveal diffusive spin transport in different model compounds of the $S = 1/2$ AFHC [23–25]. In this work we present experimental evidence for ballistic spin transport in SrCuO$_2$ at low temperature $T \ll J$ and finite fields $H \ll J$ based on $\mu$SR experiments.

The structure of SrCuO$_2$ contains chains running along the crystallographic $c$-axis, built of corner-sharing CuO squares and reminiscent of the CuO layers of cuprate superconductors. In SrCuO$_2$, two chains are joined by sharing their edges, forming a zig-zag chain. The antiferromagnetic coupling between nearest neighbors (NN) is large compared to the ferromagnetic exchange coupling $J \approx 220$ K between diagonal Cu spins [26]. Hence, it can also be described as a $S = 1/2$ chain with ferromagnetic NN and antiferromagnetic next-nearest neighbor (NNN) interactions. The resulting weak frustration can produce essential features in the behavior of the spin chain. Inelastic neutron scattering data (INS), however, indicates that both chains are decoupled and show no features associated with frustration [1]. Neutron diffraction experiments revealed anisotropic spin freezing below $\approx 5$ K in SrCuO$_2$ [27]. This frozen state is also detected by $\mu$SR, but only below $\approx 2$ K [7]. Thurber and coworkers studied the dynamics of the $q = 0$ modes in SrCuO$_2$ by $^{17}$O nuclear magnetic resonance (NMR) [24]. They found a magnetic field $B_0$ dependence of the spin–lattice relaxation rate $1/T_1 \sim B_0^n$ that is consistent with diffusive spin transport, i.e., with $n = 0.5$ [24].

In this study we used a single crystal of SrCuO$_2$ grown by the traveling solvent floating zone technique from high-purity (99.99%) precursors. The thermal transport properties of this sample have been reported by Hlubek and coworkers in [19]. A single crystal was oriented and mounted with the crystallographic $b$-axis along the muon beam direction. $\mu$SR experiments were conducted using a $^4$He flow cryostat at the GPS instrument of the Paul Scherrer Institute, Switzerland. In a $\mu$SR experiment, nearly 100% spin-polarized muons are implanted into the sample, one at a time. In cuprate materials the positively charged $\mu^+$ usually form a $\approx 1$ Å long bond with an oxygen ion, where they act as magnetic microprobes [28–30]. In a non-magnetic material the muon spins dephase, i.e., the 100% initial spin polarization $P(t)$ decays as a function of time $t$ due to the random orientation of nuclear magnetic dipole fields. The nuclear dipole field distribution can be modeled by an isotropic Gaussian distribution with width $\Delta$. This is well known and can be described by the so-called Kubo–Toyabe function $G(t, \Delta, B_0)$, which, in a longitudinal magnetic field $B_0$ has been described by Hayano [31]. Typically, $B_0 \approx 5–10$ mT is sufficient to decouple the muon spin from the nuclear dipole field distribution.

The quantum spin fluctuations of the $S = 1/2$ AFHC cause an additional independent relaxation mechanism for the muon spin ensemble. Rapid fluctuations cause an exponential relaxation of the muon spin polarization $P(t) \propto \exp(-\lambda t)$, with the spin–lattice relaxation rate $\lambda$. The overall relaxation function is then the product $P(t) = G(t, \Delta, B_0) \exp(-\lambda t)$. In our experiments, $\lambda$ is of the order of 0.01 $\mu$s$^{-1}$, and we study its field and temperature dependence as shown in figures 2 and 3. No signature of muon diffusion has been found; hence $\Delta = 0.085(2) \mu$s$^{-1}$ is temperature and field independent. Typical $\mu$SR time spectra and best fits to the data are shown in figure 1.

For diffusive spin transport in one dimension $\lambda \propto B_0^{0.5}$ is expected [32, 17, 18]. Its divergence for $B_0 = 0$ can be cut off by 3-d diffusion/coupling or by anisotropy, e.g., dipolar intrachain coupling. A useful description of experimental data has to include this cut-off for low fields/frequencies [33]:

$$\lambda(B_0) = \lambda_0 \left(1 + \sqrt{1 + (B_0/2B_c)^2}\right)^n. \tag{1}$$

![Figure 1. Typical $\mu$SR time spectra of SrCuO$_2$ at a temperature of 75 K, measured for different longitudinal magnetic fields. Solid lines are best fits to the data (see text).](image1)

![Figure 2. The field dependence of the relaxation rate $\lambda(B_0)$, here shown for $T = 75$ K, follows a clear power law behavior for $B_0 > B_c = 0.23(3)$ mT and saturates for smaller fields (see figure B.1 in appendix B for additional experimental data). The solid line is a fit of equation (1) to the data ($B_c$ was optimized simultaneously for all temperatures). The large deviation from the expected field dependence for $B_0 = 10$ mT is due to the limited time window of the experiment of approximately 12 $\mu$s. The open symbol indicates $\lambda(B_0 = 0)$.](image2)
the muon as an impurity we compare our data with recent e.g., phonons. is limited, i.e., rendered diffusive by extrinsic perturbations, $S_{\text{DSF}} \propto \int dq dq' \delta(q-q') F(q, \omega)$, with $\gamma_F$ the gyromagnetic ratios of electron (nuclear) spins, $F(q, \omega)$ and $F^\perp(q, \omega)$ are components of the hyperfine form factors parallel and perpendicular to the external magnetic field, and $S^z(q, \omega)$, $S^\perp(q, \omega)$, and $S^\parallel(q, \omega)$ are the components of the tensor of the dynamic structure factor (DSF) of electron spins, also parallel and perpendicular to the external field. In general, the muon relaxation rate $\lambda$ differs from the nuclear magnetic resonance rate $1/T_1$ only by the form factor.

A detailed knowledge of the form factor is not required because generally no ‘filtering’ of the DSF by the form factor is expected in cuprate materials. This is due to the low symmetry of the muon site, which is usually at a distance of $\approx 1 \text{ Å}$ from an oxygen ion [28–30].

The experimentally found $\lambda_{\text{NMR}} \propto 1/B_0^2$, with $n = 0.9(3)$, is in clear disagreement with the model for spin diffusion ($n = 0.5$). This leaves us with two questions: (1) is the spin transport ballistic?, and (2) how can we understand the observed power law? In the following we show that the measured power law with $n \approx 1$ can be understood by the spin excitation spectrum of the ground state of the $S = 1/2$ AFHC. In the ground state, exact calculations show that the spin transport is ballistic (see e.g. [17, 18] and references therein). Therefore, $n \approx 1$ proves ballistic spin transport in the $S = 1/2$ AFHC material SrCuO$_2$ because we find $n = 0.9(3)$ by experiment.

As a starting point, we chose a model for the ground state spin excitation spectrum that was confirmed experimentally for SrCuO$_2$ by inelastic neutron scattering [11]. This well-known model was given by Müller et al and is often used to analyze inelastic neutron scattering data [11, 36]. In this model $S^z \propto 1/\omega$, in agreement with our experimental result. We describe this model below.

According to the conjecture of Müller et al, the ground state DSF of the $S = 1/2$ AFHC is determined by the 2-spinon continuum [9–11]. In the absence of the magnetic field it has a lower bound $\epsilon_1(q) = \pi J/2 |\sin q|$ and an upper bound $\epsilon_2(q) = \pi J \sin q/2$. The contribution to the ground state DSF of the 2-spinon continuum of the $S = 1/2$ AFHC can be written as

$$S^z_2(q, \omega) = A_2 \frac{\Theta(\omega - \epsilon_1(q))\Theta(\epsilon_2(q) - \omega)}{\sqrt{\omega^2 - \epsilon_1^2(q)}},$$

(3)

where $\gamma_F(N)$ are the gyromagnetic ratios of electron (nuclear) spins, $F(q, \omega)$ and $F^\perp(q, \omega)$ are components of the hyperfine form factors parallel and perpendicular to the external magnetic field, and $S^z(q, \omega)$, $S^\perp(q, \omega)$, and $S^\parallel(q, \omega)$ are the components of the tensor of the dynamic structure factor (DSF) of electron spins, also parallel and perpendicular to the external field. In general, the muon relaxation rate $\lambda$ differs from the nuclear magnetic resonance rate $1/T_1$ only by the form factor.

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where $A_2$ is a constant, and $\Theta(x)$ is the Heaviside step function (see [37, 38] for the optimized value of $A_2$).

In the absence of a magnetic field the DSF is isotropic, and at $\omega \to 0 \omega \ll J$ two points of the reciprocal space, $q = 0$ and $\pi$, contribute mostly to the relaxation rate (2) [9–11]. On the other hand, a nonzero external magnetic field introduces an anisotropy for the components of the dynamic correlation functions of the $S = 1/2$ AFHC, shifting the contributing points away from $q = 0, \pi$ (see appendix A). However, in our $\mu$SR experiments the external magnetic field was much smaller than the exchange constant along the spin chain, and this shift is negligible. In $\mu$SR experiments we have $\omega = \omega_0 = \gamma \mu B_0$. In the ballistic regime, according to equation (3), the relaxation rate in the ground state has to be inversely proportional to the value of the external field

$$\lambda \propto \frac{1}{B_0}. \quad (4)$$

Notice that the value $A_2$ is field dependent [39–43]. At low fields, this can cause a weak logarithmic dependence of the relaxation rate in addition to the inverse proportionality (4). The power law (4), $\lambda \propto B_0^{-n}$ with $n = 1$ valid for the ground state DSF, is in agreement with the experimentally found power law with $n = 0.9(3)$. According to above arguments, this is evidence for ballistic spin transport in SrCuO$_2$. In the following we discuss the well-known effects of exchange anisotropy, temperature and next-nearest neighbor (NNN) exchange interaction on the power law (4).

Uniaxial anisotropy of the exchange interaction can alter the frequency dependence of $S^{zz}(q, \omega)$ and hence the exponent in the power law (4):

$$S^{zz}(q, \omega) = A \frac{\Theta(\omega - \epsilon_1(q))\Theta(\epsilon_2(q) - \omega)}{(\omega^2 - \epsilon_1^2(q))^{\alpha_0}(\epsilon_2^2(q) - \omega^2)^{(1/2) - \alpha_0}}. \quad (5)$$

The exponent $\alpha_0$ can be calculated from the finite size corrections to the ground state energy and $\alpha_0 \approx 1 - Z^2 = (\pi - 2\eta)/(2\pi - 2\eta)$, where $Z = \sqrt{\pi/2(\pi - \eta)}$ is the dressed charge of the spin chain model [44] in zero external magnetic field. The parameter $\eta$ is related to the magnetic anisotropy of the exchange interaction $\cos q = J'/J$ for easy-plane anisotropy, and $\cosh q = J'/J$ for easy-axis anisotropy of the spin–spin interaction along the chain ($A$ is $\eta$-dependent). Hence, our experimental results permit a small anisotropy. However, an exact determination of $\eta$ is limited by the experimental accuracy.

In our experiments temperatures were small compared to the intrachain exchange interactions, $T \ll J, J'$. For low fields we can then estimate the temperature dependence of the relaxation rate $\lambda$ from the temperature dependence of the DSF by conformal field theory [44]:

$$\lambda \propto \frac{1}{B_0^2} \left(\frac{2\pi aT}{v}\right)^\gamma \quad (6)$$

where $\alpha$ is the cut-off parameter, $v = \pi J \sin(\eta)/2\eta$ is the spinon velocity ($v \approx \pi J/2$ for weak anisotropy) and $\gamma$ is related to the Luttinger liquid exponent, connected to the dressed charge. For the isotropic $S = 1/2$ AFHC we have $\gamma \approx 0$. The temperature dependence of $\lambda_0$ in figure 2 is constant below approximately 150 K, hence it is in agreement with $\gamma \approx 0$.

Formally the double chain in SrCuO$_2$ can be treated by introducing weak NNN exchange interaction. The main effect of the then frustrated NN and NNN exchange interaction is an additional minimum (maximum) for $q = \pi/2$ in the lower (upper) boundary of the DSF [45]. However, as we pointed out above, the main contributions to the $\mu$SR (and NMR) spin–lattice relaxation rate have to come from $q = 0, \pi$, which are not essentially changed by spin frustration in the chain. In addition, it is possible that the low temperature exponent $\gamma$ becomes nonzero due to spin frustration [46]. This effect should be small, because we experimentally find $\gamma \approx 0$ (see above). Furthermore, any effects of the NNN on the DSF would be detected by inelastic neutron scattering (INS). The absence of any such effect on the DSF measured by INS [1] shows that, up to energies of the order of the exchange interaction $J \approx 2100$ K, the NNN interactions have negligible effects.

It turns out that the results of Monte-Carlo simulations [15, 16] indicate diffusive behavior of the relaxation rate in the $S = 1/2$ AFHC. Those results are valid for high and intermediate temperatures [15, 16], i.e., $T \gg J$, or $T \approx J$. In our situation we have $T \approx 75$ K, and $J \approx 2000$ K, i.e., $T \ll J$, and we cannot apply the results of Grossjohann et al [15, 16] for the explanation of our experiments. Also, similar diffusive behavior was predicted within the field-theoretical approximation [17, 18]. However, the perturbation scheme that Sirker et al use in [17, 18] for the calculation of the self-energy (mass operator) cannot be applied in our case, because both the perturbation and the main part of the Hamiltonian are determined by the same exchange constant $J$, i.e., formally there is no small parameter in our situation. The real part of the calculated self-energy defines the renormalization of the excitation spectrum of the low-energy quasiparticle. On the other hand, the imaginary part, which is connected with $\gamma$, determines the damping of that quasiparticle (or the inverse lifetime of that excitation). For the conditions of our experiment ($T = 75$ K and $J = 2100$ K) we can estimate $\gamma(T = 75$ K) $\approx 4.5$ (in energy units $h = 1$). In [17, 47] the damping of low-energy spin excitations $\gamma$ is found to be independent of $q$. Such a large $\gamma$ implies that at the temperatures of our experiment in SrCuO$_2$ all low-energy excitations have to decay (at least for small enough values of quasimomenta $q$). It is, in fact, the manifestation of the difference between ballistic and diffusive transport: in the ballistic regime the transport is determined by the dynamics of low-energy excitations, while in the diffusive regime all such excitations are ‘smeared out’ due to their damping, and transport properties are described by the hydrodynamics. In that context, it is difficult to believe that in SrCuO$_2$ low-energy excitations do not exist at $T = 75$ K $\ll J$. It is known that low-energy excitations in SrCuO$_2$ were observed by the neutron scattering technique for $T \approx 12$ K [27, 1], and by ARPES for $T \approx 300$ K [48], which contradicts the large damping of low-energy excitations proposed in [17, 47].
Recently, Lancaster et al. [49] also observed ballistic spin transport in \( \text{Rb}_2\text{Cu(MO}_4\text{)}_3 \), an organic \( S = 1/2 \) AFHC compound with \( J = 10 \) K. They reported a logarithmic field dependence of the muon spin–lattice relaxation rate, characteristic for isotropic spin chains. The power law field dependence we report in the present work, on the other hand, implies that the magnetic exchange interaction along the spin chains in \( \text{SrCuO}_2 \) is slightly anisotropic. The different temperature ranges in our study and in [49] are related to different values of the exchange integrals.

Concerning the differences between our results and previous published results [24, 23, 25] on \( S = 1/2 \) AFHC model compounds, we only want to mention the importance of impurities. Hammerath et al. have recently shown [34] that in \( \text{SrCuO}_2 \), bond disorder leads to the formation of a gap in the spin excitation spectrum at low temperatures. This is not fully understood, but it follows that in experimental studies that probe the low-energy spin excitations, impurities play a crucial role because they can alter the low-energy excitation spectrum away from that of the \( S = 1/2 \) AFHC. In most cases this circumstance is not as obvious, because no gap is observed. However, it is clear that impurities can change the low-energy excitation spectrum of the \( S = 1/2 \) AFHC drastically. The samples that we have studied are of high purity, as has been shown by heat transport experiments [19]. They demonstrated that the mean free path of magnetic heat conduction is of the order of 1 \( \mu \text{m} \) [19]. This is exceptionally high and can only be due to the high quality of our samples [19]. We are therefore led to believe that the apparent differences between our results and previous results [24, 23, 25] may be due to the very low amount of impurities in our samples.

In summary, the magnetic field dependence of the muon spin–lattice relaxation rate \( \lambda \) in \( \text{SrCuO}_2 \) is \( \lambda \propto B_0^n \), with \( n = 0.9(3) \). This is in close agreement with \( n = 1 \), which follows from the dynamic structure factor expected for the ground state of the isotropic antiferromagnetic \( S = 1/2 \) Heisenberg chain (AFHC). We conclude that in this system the low-energy spin dynamics is determined by the eigenstates of the Heisenberg spin chain Hamiltonian. Therefore, in \( \text{SrCuO}_2 \), spin transport is ballistic at low temperatures and fields 0 < \( H, T \ll J \). Furthermore, the absence of a temperature dependence for \( T \lesssim 150 \) K of the spin–lattice relaxation rate \( \lambda(0) = \lambda_0 \) in zero field indicates that frustration due to NNN interaction does not influence the low-energy spin dynamics. In addition it shows that spin and heat transport [19] are decoupled in this system. This does not contradict the often cited magnetic contribution to the heat conductivity of low-dimensional magnets. It follows that the mechanism causing the temperature dependence of the magnetic heat transport is not relevant for the spin transport in \( \text{SrCuO}_2 \).

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Appendix A

In this appendix we carefully show why the main contributions to the dynamical structure factor come from the points \( q = 0, \pi \) of the dispersion law \( \varepsilon(q) \).

The muon or NMR spin–lattice relaxation rate \( T_1^{-1} \) can be presented as [35]

\[
\frac{1}{T_1} = \frac{\gamma \hbar^2}{2} \lim_{\omega \to \omega_L} \sum_q (F^z(q)S^z(q, \omega)) \quad (A.1)
\]

\[ + \frac{1}{2} F^\pm(q)[S^\pm(q, \omega) + S^\mp(q, \omega)], \quad (A.2) \]

where \( \gamma \text{e(N)} \) are the gyromagnetic ratios of electron (nuclear) spins (\( \gamma_e = g \mu_B \)), \( \omega_L \) is the Larmor frequency of nuclear spins, \( F^z(q) \) and \( F^\pm(q) \) are the hyperfine form factors of nuclear spins, parallel and perpendicular to the external dc magnetic field, and \( S^z(q, \omega) \), \( S^\pm(q, \omega) \), and \( S^\mp(q, \omega) \) are the components of the tensor of the dynamical structure factor of electron spins, also parallel and perpendicular to the external field. The limit \( \omega \to \omega_L \approx 0 \) is taken because the resonance frequency of nuclear spins is several orders of magnitude smaller than the frequencies corresponding to the characteristic magnetic energy scales of the electronic spin system.

The ground state and low temperature behavior of the dynamical structure factor (dynamical correlation function) of the Heisenberg spin-1/2 chain in the external magnetic field have been studied in [11]. It was pointed out there that several continua of low-energy gapless excitations of the Heisenberg chain (nowadays they are known as spinons, [50] and the continua, studied in [11] are now known as two-spinon continua) yield the main contribution to the dynamical structure factor. That conjecture [11], supported by their numerical calculations for small-size finite spin chains, was later confirmed by a calculation, which used exact Bethe ansatz equations in the ground state of the Heisenberg chain [8]; it was shown that about 75% of the dynamical structure factor of the antiferromagnetic Heisenberg chain is determined by the two-spinon continuum.

In the absence of the magnetic field the dynamical structure factor is isotropic, and at \( \omega \to \omega_L \) two points of the reciprocal space, namely, \( q = 0 \) and \( \pi \), contribute mostly to the spin–lattice relaxation rate. On the other hand, the nonzero external magnetic field introduces anisotropy for the components of the dynamical correlation functions of the Heisenberg chain. It was shown [11] that for \( B_0 \neq 0 \) the main contribution to the longitudinal component of the dynamical structure factor \( S^z(q, \omega) \) of the spin chain is determined by
the lower and upper boundaries of the two-spinon continuum, defined as (we use the numeration for branches from [11])

$$
\varepsilon_{2L} = 2D \sin(q/2) \cos[(q + 2\pi m)/2],
$$

$$
\varepsilon_{2U} = 2D \sin(q/2) \cos[(q - 2\pi m)/2],
$$

where \(0 \leq q \leq \pi\), \(D = (1/4)(2\pi J - H(\pi - 2))\), \(m = (1/\pi)\sin^{-1}(H/2D)\) is the magnetization of the spin chain per site for \(0 \leq H \leq H_s\), where \(H = \gamma \hbar B_0\), and \(H_s = 2J\) is the quantum critical point for the Heisenberg spin chain, at which the chain undergoes the quantum phase transition to the spin-polarized state (with \(m = 1/2\)). This continuum for \(H \to H_s^+\) collapses into a single branch of the magnon with a negligible weight. Upper and lower branches of that two-spinon continuum are presented in figure A.1 for \(J = 1\) and \(H = 0.5J\).

One can see that the nonzero magnetic field causes the shift of the most important contribution for \(S^z(q, \omega = 0)\) from \(q = \pi\) to \(q = \pi(1 - m)\), while the point \(q = 0\) remains important. Let us call those relevant points in the \(\omega-q\) space as points P1 for \(\omega = 0, q = 0\), and P2 for \(\omega = 0, q = \pi(1 - 2m)\).

As for the transverse components, they are mostly determined by three continua. Namely, for \(S^{-+}(q, \omega)\) the lower and upper boundaries are

$$
\epsilon_{3L} = 2D \sin(q/2) \cos[(q - 2\pi m)/2] - H, \\
\epsilon_{3U} = 2D \sin(q/2) - H,
$$

where \(2\pi m \leq q \leq \pi\). For nonzero magnetic field this contribution persists, but its range shrinks with the growth of field, vanishing at \(H = H_s\). For \(S^-(q, \omega)\) the first two-spinon continuum has the following lower and upper boundaries

$$
\epsilon_{5L} = 2D \sin(q/2) \cos[(q + 2\pi m)/2] + H, \\
\epsilon_{5U} = 2D \sin(q/2) \cos[(q - 2\pi m)/2] + H,
$$

where \(0 \leq q \leq \pi\). The contribution of this continuum was considered [11] to be relevant only for finite chains, and has to vanish in the thermodynamic limit \(L \to \infty\), where \(L\) is the number of sites in the chain. Finally, the contribution to \(S^+(q, \omega)\) comes also from the continuum with the upper and lower boundaries

$$
\epsilon_{6L} = 2D \sin[(\pi - q)/2] \cos[(\pi - q + 2\pi m)/2], \\
\epsilon_{6U} = 2D \sin[(\pi - q)/2] \cos[(\pi - q - 2\pi m)/2],
$$

where \(0 \leq q \leq \pi\). This continuum is essential for \(H \leq H_s\), and dominant for \(H \geq H_s\), where elementary excitations (magnons) are gapped. Figure A.1 illustrates the two-spinon continua for the spin-1/2 antiferromagnetic Heisenberg chain, relevant for the transverse components of the dynamical structure factor, for \(H = 0.5J\) and \(J = 1\).

We see that for the \(S^+(q, \omega = 0)\) and \(S^-(q, \omega = 0)\) components the point \(q = \pi\) remains relevant, while instead of \(q = 0\) the main contribution comes from \(q = 2\pi m\), opposite to the situation with the longitudinal components. Let us call those relevant points in the \(\omega-q\) space as points P3 for \(\omega = 0, q = 2\pi m\), and P4 for \(\omega = 0, q = \pi\).

Such a magnetic field behavior mostly determines the dynamics of the Heisenberg antiferromagnetic spin chain at frequencies \(\omega \sim J/h\). For higher frequencies the contributions from the four-spinon continuum become more relevant [8].

Now let us turn to the conditions of our experiment in \(\text{SrCuO}_2\). Here the exchange energy \(J \sim 2100\) K, and in \(\mu\)SR experiments the value of the magnetic field was between zero and 10 mT. Then it follows that in the above formulas we can neglect \(m \ll 1\), and the main contributions to the integral come from the points \(q = 0\) and \(\pi\). The contributions from other points in the momentum space are exponentially small and can be neglected (a similar neglect of the contributions from other points was used in many theoretical and experimental works that studied low-frequency dynamical properties of the antiferromagnetic spin-1/2 chains in an external magnetic field, see, e.g., [51–53]). Notice that the contributions from the four-spinon and higher-spinon continua to the low-energy dynamical structure factor at \(\omega \sim 0\) also come from the points \(q = 0, \pi\).

### Appendix B

In this appendix we show additional experimental data in figure B.1.
Figure B.1. Muon spin–lattice relaxation rate $\lambda$ of SrCuO$_2$ as a function of applied magnetic field $B_0$ at various temperatures $T$. Lines are fits to the data, as described in the main part of the paper. Measurements for $B_0 < 2$ mT were only performed at selected temperatures.

References

[1] Zaliznyak I A et al 2004 Phys. Rev. Lett. 93 087202
[2] Imada M, Fujimori A and Tokura Y 1998 Rev. Mod. Phys. 70 1039–263
[3] Maekawa S and Tohyama T 2001 Rep. Prog. Phys. 64 383
[4] Sachdev S and Keimer B 2011 Phys. Today 64 (2) 29–35
[5] Lee P A, Nagaosa N and Wen X-G 2006 Rev. Mod. Phys. 78 17–85
[6] Motoyama N, Eisaki H and Uchida S 1996 Phys. Rev. Lett. 76 3212–5
[7] Matsuda M et al 1997 Phys. Rev. B 55 R11953–6
[8] Caux J-S and Maillet J M 2005 Phys. Rev. Lett. 95 077201
[9] Müller G et al 1979 Phys. Rev. Lett. 43 75–8
[10] Müller G et al 1981 J. Phys. C: Solid State Phys. 14 3399
[11] Müller G et al 1981 Phys. Rev. B 24 1429–67
[12] Žnidarič M 2011 Phys. Rev. Lett. 106 220601
[13] Prosen T 2011 Phys. Rev. Lett. 106 217206
