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Spatiotemporal crossover between low- and high-temperature dynamical regimes in the quantum Heisenberg magnet

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The stranglehold of low temperatures on fascinating quantum phenomena in one-dimensional quantum magnets has been challenged recently by the discovery of anomalous spin transport at high temperatures. Whereas both regimes have been investigated separately, no study has attempted to reconcile them. For instance, the paradigmatic quantum Heisenberg spin-1/2 chain falls at low-temperature within the Tomonaga-Luttinger liquid framework, while its high-temperature dynamics is superdiffusive and relates to the Kardar-Parisi-Zhang universality class in $1 + 1$ dimensions. This work aims at reconciling the two regimes. Building on large-scale matrix product state simulations, we find that they are connected by a temperature-dependent spatiotemporal crossover. As the temperature T is reduced, we show that the onset of superdiffusion takes place at longer length and time scales $\propto 1/T$. This prediction has direct consequences for experiments including nuclear magnetic resonance: it is consistent with earlier measurements on the nearly ideal Heisenberg $S = 1/2$ chain compound Sr_2CuO_3 yet calls for new and dedicated experiments.

Introduction.— At low temperatures, reduced spatial dimensionality greatly enhances quantum fluctuations in physical systems, giving rise to exotic properties. In that regard, one-dimensional (1D) quantum many-body systems have always been influential and generically fall into two classes [1, 2]: on the one hand, gapless low-energy excitations described in the framework of Tomonaga-Luttinger liquid (TLL), and on the other, a gapped behavior. Theoretical predictions have been intensively checked by experiments in various contexts, ranging from ultra-cold atom setups to quantum magnets [3, 4].

At energy $\hbar\omega \ll k_B T$, the physics is usually thought of in terms of thermal rather than quantum effects. This regime had not been thought to hold phenomena as compelling as its low-temperature counterpart until very recently. Indeed, recent theoretical progress suggests that the equilibrium and out-of-equilibrium dynamics of some 1D quantum systems can exhibit peculiar behaviors and contain information about the intrinsic quantum features, even at very high temperatures [5–7].

While such many-particle systems are governed at the microscopic level by the Schrödinger equation, they display in the long-time and long-wavelength limits an emergent coarse-grained hydrodynamic behavior. An analogy can be made with classical fluid dynamics: one does not describe individual particles with Newton’s laws of motion but relies instead on phenomenological continuous differential equations, ideally more amenable. The derivation of hydrodynamic equations is based essentially on continuity equations of conserved quantities (e.g., mass, energy, etc.), assuming local equilibrium [8].

Quantum systems also possess conservation laws, and depending on those, one expects the emergence of different kinds of coarse-grained hydrodynamic descriptions. Singularly in 1D, a class of quantum systems – known as integrable – has an infinite set of nontrivial conserved quantities which can lead to anomalous dynamical behaviors [5–7, 9–34].

Integrable systems are typically described by very fine-tuned models but some of them can be reliably realized in the lab (e.g., the Lieb-Liniger model representing a gas of

one-dimensional bosons with contact repulsion [35, 36]) and found with high fidelity in nature (e.g., the spin-1/2 Heisenberg chain of magnetic moments coupled by a nearest-neighbor exchange interaction [2]). In that context, some of the theoretical predictions have been successfully tested on 1D cloud of trapped ^{87}Rb [37, 38] and ^7Li [39] atoms for out-of-equilibrium dynamics and by neutron scattering on the quantum magnet KCuF_3 at thermal equilibrium [34].

In the case of quantum magnets, it has been numerically conjectured, based on microscopic simulations, that in the limit of infinite temperature, the spin dynamics of the $S = 1/2$ Heisenberg chain is anomalous and belongs to the Kardar-Parisi-Zhang (KPZ) universality class in $1 + 1$ dimensions [20, 40]. It is characterized by a dynamical exponent $z = 3/2$, controlling the length-time scaling of the dynamical properties. This exponent has been recently observed in the high-temperature neutron spectrum of KCuF_3 [34], which is directly proportional to the dynamical structure factor, probing spin-spin correlations.

Here, we seek to reconcile the low-temperature physics of the $S = 1/2$ Heisenberg chain, falling within the gapless TLL category, with the recently found infinite-temperature KPZ hydrodynamics. Whereas both regimes have been studied independently, no work has attempted to bring them together. In this Letter, we precisely define the long-time and long-wavelength limits for the emergence of anomalous dynamics versus the temperature. We find that these limits define a spatiotemporal crossover beyond which hydrodynamics take place. As the temperature is lowered, the crossover is pushed towards infinity and eventually disappears at exactly zero temperature, see Fig. 1. This scenario allows one to recover the well-known zero temperature results where KPZ hydrodynamics is absent. Moreover, because experimental dynamical condensed matter probes such as neutron scattering or nuclear magnetic resonance (NMR) work for all practical purposes at a finite frequency and finite temperatures, it is paramount to better understand and quantitatively define the theoretical limits. We discuss the implication of our results for experiments

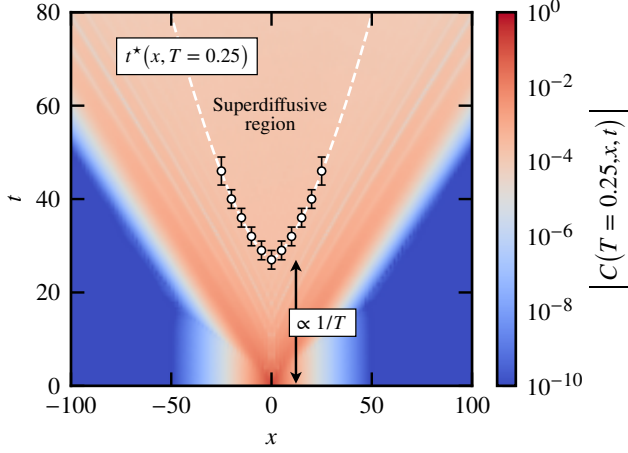


FIG. 1. Log-scale intensity plot of the Euclidean norm of the spin-spin correlation (2) at $T = 0.25$. Simulation obtained for $L = 256$ with $\chi = 1024$. The goal of this work is to determine and study the superdiffusive region delimited by the spatiotemporal crossover t^* of Eq. (3) versus the temperature (white circles and dashed white line). As the temperature is decreased, we find that the superdiffusive region is shifted vertically to longer and longer times by a factor $\propto 1/T$, and eventually disappear at exactly zero temperature.

and confront our findings to earlier high-temperature NMR experiments on the nearly ideal Heisenberg spin-1/2 compound Sr_2CuO_3 [41].

Model and method.— The 1D spin-1/2 Heisenberg model is described by the lattice Hamiltonian,

$$\hat{\mathcal{H}} = J \sum_j \hat{\mathbf{S}}_j \cdot \hat{\mathbf{S}}_{j+1}, \quad (1)$$

with $\hat{\mathbf{S}}_j = (\hat{S}_j^x, \hat{S}_j^y, \hat{S}_j^z)$ and $J > 0$ the nearest-neighbor antiferromagnetic exchange. To investigate the thermal equilibrium spin dynamics, we consider the time-dependent spin-spin correlation function,

$$C(T, x, t) = \text{tr}(\hat{\mathbf{S}}_x(t) \cdot \hat{\mathbf{S}}_0(0) \hat{\rho}_T) \in \mathbb{C}, \quad (2)$$

with $\hat{\rho}_T = e^{-\hat{\mathcal{H}}/k_B T} / \text{tr}(e^{-\hat{\mathcal{H}}/k_B T})$ the thermal density matrix of the system at temperature T and $\hat{\mathbf{S}}_j(t) = e^{i\hat{\mathcal{H}}t/\hbar} \hat{\mathbf{S}}_j e^{-i\hat{\mathcal{H}}t/\hbar}$ the time-dependent spin operator in the Heisenberg picture. We set $J = k_B = \hbar = 1$ in the following. We compute the correlation function (2) based on a numerical matrix product state (MPS) approach [42, 43] where we represent the mixed state as a pure state in an enlarged Hilbert space [44, 45]. We use the time-evolving block decimation algorithm [46] along with a fourth-order Trotter decomposition [47] to handle the exponential operators [48]. To ensure convergence of the numerical data, we study in the Supplementary Information (SI) the effect of the bond dimension χ of the MPS, which is the control parameter of the simulations (larger is better, but computationally more expensive) [49].

At fixed distance x and temperature T , the hydrodynamics regime is characterized by an algebraic decay of the Euclidean

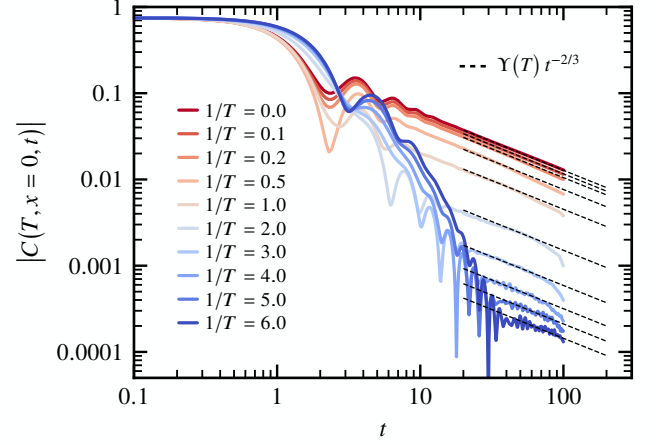


FIG. 2. Time dependence of the norm of the spin-spin correlation (2) at $x = 0$ for various temperatures T . Simulations obtained for $L = 256$ with $\chi = 1024$. At long time, it displays an algebraic decay with time, according to Eq. (3). It is well-fitted by the form $\Upsilon(T) t^{-2/3}$ with $\Upsilon(T)$ a temperature-dependent prefactor decreasing with the temperature reported in Fig. 3(b). The deviation from the genuine power-law at long-time is the result of the bond dimension being too small [49].

norm of the spin-spin correlation (2) function at long time,

$$|C(T, x, t)| \propto t^{-1/z} \quad \text{for } t \gtrsim t^*(x, T), \quad (3)$$

with z the dynamical exponent. The long-time limit is denoted by the crossover time t^* which we aim to identify, see Fig. 1. Depending on the microscopic model, three values for the exponent z have been reported for 1D quantum magnets: $z = 3/2$ corresponding to superdiffusion, $z = 1$ for ballistic and $z = 2$ for diffusion [24, 25]. Superdiffusion is expected for the isotropic spin-1/2 Heisenberg model of Eq. (1).

Autocorrelation.— We first consider the autocorrelation function ($x = 0$) versus time for different temperatures, as plotted in Fig. 2. Two regimes are clearly visible, delimited by the crossover time $t^*(x = 0, T)$ [49]. Beyond the crossover time and for all temperatures, one finds the expected power-law decay $\propto t^{-2/3}$ of superdiffusive hydrodynamics. Note that the rapid change of slope from the genuine power-law, at the longest times displayed, is the result of the bond dimension being too small and not a physical effect [49].

With high-temperature physics beyond t^* , one can suspect low-temperature features at shorter times. For instance, the oscillating behavior observed in the norm of the autocorrelation is reminiscent of a change of sign in the real and imaginary part [49], signaling antiferromagnetic correlations as the temperature is lowered. The long-time asymptotic of $C(T = 0, x = 0, t)$ have been studied at exactly zero temperature [50, 51]. It is composed by several power-law decaying contributions with the slowest one being $\propto t^{-1}$ (up to logarithmic corrections inherent to the isotropic spin-1/2 Heisenberg antiferromagnet [49, 52–59]). We cannot identify this regime in Fig. 2, which we attribute to insufficiently low temperatures, see the SI for additional data [49].

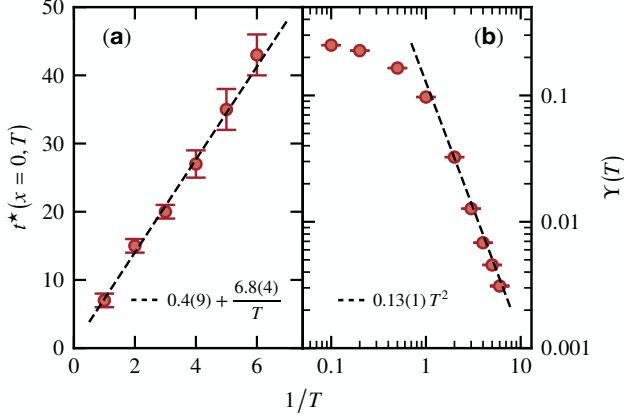


FIG. 3. The data points are extracted from Fig. 2. (a) Temperature dependence of the crossover timescale $t^*(x=0, T)$ beyond which the algebraic decay $\propto t^{-2/3}$ for superdiffusive hydrodynamics emerges, see Eq. (3). It shows a linear dependence with the inverse temperature (dashed line). (b) Temperature dependence of the prefactor $\Upsilon(T)$ of the algebraic decay $\propto t^{-2/3}$ for superdiffusive hydrodynamics. At low temperatures $T \lesssim 1$, it follows a quadratic dependence $\propto T^2$ (dashed line).

We now turn our attention to the temperature dependence of the crossover time $t^*(x=0, T)$. It is plotted in Fig. 3(a) versus the inverse temperature and shows a linear dependence. It can be understood as follows. It is well-known that a finite temperature induces a thermal correlation length ξ which diverges as $T \rightarrow 0$ as $\propto u/T$ (up to logarithmic corrections [49, 53]) with u the velocity of low-energy excitations in the spin-1/2 chain. Moreover, the dynamical correlation function (2) can also be thought of as measuring the spreading of a spin excitation. In this picture, the system behaves like a TLL for $t \lesssim \xi/u$, which can be identified as the crossover time $t^*(x=0, T) \propto 1/T$. Hence, the onset of superdiffusive hydrodynamics simply takes place as the low-energy physics gets suppressed by the finite temperature. It is only at zero temperature that the system is strictly critical and thus does not display any sign of anomalous high-energy dynamics. In addition to the linear dependence with $\propto 1/T$, there is an $O(1)$ constant in Fig. 3(a) which coincides with the very short-time dynamics where $|C(T, x=0, t \approx 0)| \approx 0.75$.

At infinite temperature, it has been established that the dynamics belong to the 1+1 KPZ universality class [20, 40], as it shows the same scaling laws as appear in the KPZ equation itself: $\partial_t h = \frac{1}{2}\lambda(\partial_x h)^2 + \nu\partial_x^2 h + \sqrt{\sigma}\eta$ with $h \equiv h(x, t)$, $\eta \equiv \eta(x, t)$ a normalized Gaussian white noise, and λ, ν, σ parameters. It is a Langevin equation, with no quantum roots – and which makes the observation of its physics in a quantum magnet rather puzzling. In the right limits, the noise-averaged slope correlations behave as [60, 61],

$$C_{\text{KPZ}}(x, t) \simeq \chi_s (\lambda_{\text{KPZ}} t)^{-2/3} f_{\text{KPZ}} \left[x (\lambda_{\text{KPZ}} t)^{-2/3} \right] \quad (4)$$

with $\chi_s = \sigma/2\nu$ the static spin susceptibility [49], $\lambda_{\text{KPZ}} =$

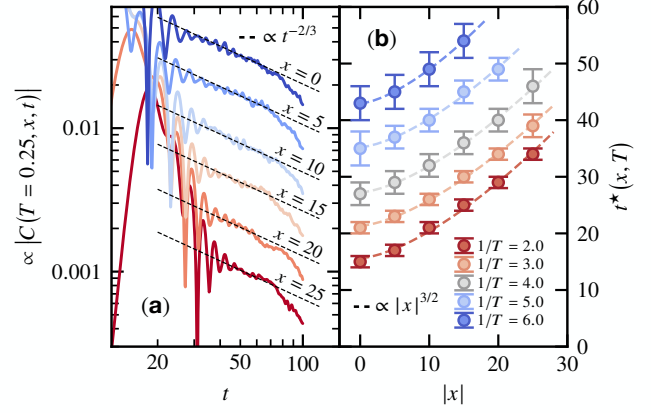


FIG. 4. (a) Time dependence of the norm of the spin-spin correlation (2) at $T = 0.25$ for various distances x . Simulations obtained for $L = 256$ with $\chi = 1024$. The curves have been shifted vertically for visibility. At long time, it displays an algebraic decay with time, according to Eq. (3), well-fitted by the form $\propto t^{-2/3}$. The deviation from the genuine power-law at long time is the result of the bond dimension being too small [49]. (b) Spatial dependence of the crossover time $t^*(x, T)$ beyond which the algebraic decay $\propto t^{-2/3}$ for superdiffusive hydrodynamics emerges, see Eq. (3). The dashed lines are fits of the form $A + B|x|^{3/2}$ with $A \equiv t^*(0, T)$ and $B = 0.17(3)$ found to be temperature-independent [49].

$\sqrt{2}\lambda$, and f_{KPZ} the KPZ scaling function [62]. The numerical observation of the scaling (4) for the Heisenberg spin chain through the spin-spin correlation (2) served as a conjecture regarding the nature of its dynamics [20]. A theoretical scenario for how KPZ hydrodynamics emerges in the Heisenberg chain has been advanced [30]. A relation between the parameters of the KPZ equation with those of the microscopic quantum model has been proposed [26]. Here, by identifying the prefactor of $C_{\text{KPZ}}(x=0, t)$ in Eq. (4) with the prefactor $\Upsilon(T)$ of the power-law decay $\propto t^{-2/3}$ shown in Fig. 3(b), we are able to report on the temperature dependence of the parameters. The high-temperature data points are compatible with Ref. 26. In addition, for $T \lesssim 1$, we find that $\Upsilon(T) = 0.13(1)T^2$, and therefore that $\chi_s \lambda_{\text{KPZ}}^{-2/3} f_{\text{KPZ}}(0) \propto T^2$. We argue in the following that this behavior is compatible with earlier NMR experiments on Sr_2CuO_3 [41, 49].

The definition of the crossover time t^* in Eq. (3) for the onset of superdiffusion is related to the power-law dependence $\propto t^{-2/3}$ and not f_{KPZ} of Eq. (4). It is well-known that unambiguously identifying the scaling function from microscopic simulations with f_{KPZ} requires great numerical precision and long-time data for all distances x [20]. This is beyond the capability of our simulations at low temperatures. Instead, we consider the spatial dependence of t^* for $|x| > 0$.

Spatiotemporal crossover.— The time-dependent spin-spin correlation function (2) is associated with a light-cone structure and we therefore expect $t^*(x, T)$ to be an increasing function with the distance $|x|$. It is verified in Fig. 4(a) where we plot its time dependence at fixed temperature ($T = 0.25$). As $|x|$ increases, the onset of superdiffusion takes place at longer

and longer times, and we display the crossover timescale in Fig. 4(b) for different temperatures. Because we can only reliably estimate it for $|x| \lesssim 30$, it is difficult to draw a definite conclusion on its scaling. Nevertheless it is compatible with a superdiffusive length-time scaling of the form,

$$t^*(x, T) = 0.4(9) + \frac{6.8(4)}{T} + 0.17(3)|x|^{3/2}, \quad (5)$$

with the first two terms obtained from the $t^*(x = 0, T)$ data, see Fig. 3(a). The prefactor of $|x|^{3/2}$ is found independent of the temperature [49]. The reported numerical parameters are obtained by least-square fitting. The spatiotemporal crossover time (5) is plotted on top of the norm of the spin-spin correlation in Fig. 1 for $T = 0.25$. Note that based on this picture, we expect logarithmic corrections for the temperature dependence, but they are not detectable from our simulations [63].

Experimental consequences.— Although we have focused on the norm of the spin-spin correlation (2), we find that $|\Im C(T, x, t)| \ll |\Re C(T, x, t)|$ for $t \gtrsim t^*$, and that the superdiffusive power-law $\propto t^{-2/3}$ only holds for the real part [49], which therefore hosts the relevant high-temperature physics. For instance, superdiffusion was observed in KCuF_3 by neutron scattering in the limit of small momentum and vanishing frequency [34], which probes the Fourier transform to momentum and frequency spaces of $C(T, x, t)$.

Another promising experimental technique for investigating high-temperature hydrodynamics is NMR, which has been successfully used to characterize the low-temperature TLL regime in numerous spin compounds [59, 64–71]. Nuclear spins are polarized via a static magnetic field (ideally weak) and then perturbed by an electromagnetic pulse of frequency ω_0 , chosen to target specific nuclei as per the Zeeman splitting. Following the perturbation, the nuclear spins relax over time with an energy transfer to the electrons. When the nuclear and electronic spins belong to the same atom, the relaxation rate is related to the autocorrelation function, $1/T_1 \sim \int_0^{1/\omega_0} \Re C(T, x = 0, t) dt$ [72–74]. With ω_0 of the order of a few mK, it usually leads to a frequency-independent $1/T_1$ as long as the correlation decays quickly enough. Here, the hydrodynamics regime should lead instead to $1/T_1 \propto \omega_0^{1/z-1}$ and give access to z in the right frequency regime. According to Eq. (5), the corresponding crossover frequency scale, $\omega^* \sim 1/t^*$, goes as $\propto T$, and superdiffusion will be visible if $\omega_0 \ll \omega^* \sim T$. Considering the experimental range of ω_0 , this condition is fulfilled even at low temperatures, where measurements are often less noisy and less subject to spoiling effects such as phonons.

Thus, the existence of a finite spatiotemporal crossover $t^*(x, T)$ in the form of Eq. (5) confirms that superdiffusive hydrodynamics is within the experimentally relevant window of parameters with respect to temperatures, time and length scales for quantities involving $\Re C(T, x, t)$.

In fact, a power-law behavior of the form $1/T_1 \propto \omega_0^{-\alpha}$ has been reported in the nearly ideal spin-1/2 Heisenberg antiferromagnet Sr_2CuO_3 ($J \simeq 2200$ K) at $T = 295$ K a couple of decades ago [41]. NMR was performed on the ^{17}O , coupled

symmetrically to the Cu^{2+} carrying the relevant electronic spin, which filtered out the $q = \pm\pi$ contributions in the $1/T_1$ due to form factors, but not the long-wavelength modes $q = 0$ holding hydrodynamics. Although the measurement accuracy was not sufficiently precise to extract the exponent α , the results are compatible with $\alpha \approx 0.33$, which corresponds to $z = 3/2$ [49]. In addition, the authors find that at fixed frequency, the NMR relaxation rate may be approximated by an empirical form $1/T_1 T \approx a + bT$ for $T \ll J$ with a and b fitting constants. When dropping a , this is compatible with $\Upsilon(T) \propto T^2$ reported in Fig. 3(b) [49], which relates to the temperature dependence of the parameters of the KPZ equation.

Today’s theoretical understanding of the dynamics of 1D quantum systems and our results call for new NMR experiments on spin chains at high temperatures. It would provide a complementary probe to neutron scattering [34] to access anomalous spin transport in quantum materials.

Conclusion.— Building on large-scale MPS calculations, we reconciled the well-established low-temperature dynamics of the quantum Heisenberg spin-1/2 chain with the recently predicted high-temperature superdiffusive regime related to KPZ hydrodynamics. We have found that both coexist, and the transition from one to the other takes the form of a spatiotemporal crossover. The crossover is controlled by the temperature: as the temperature is lowered, the growing quantum correlations between degrees of freedom push the onset of superdiffusion to longer length and time scales as $\propto 1/T$. We also reported on the temperature dependence of the parameters of the KPZ equation, which should provide useful guidance in relating them to the microscopic parameters of the quantum model. We also showed that only the real part of the spin-spin correlations holds the superdiffusive hydrodynamics. Finally, we discussed the experimental consequences of our results for condensed matter probes. We motivated NMR experiments as a great way to measure spin transport in quantum materials and showed that earlier results are compatible with the current theoretical understanding yet calling for new experiments in quantum spin chains. Because NMR requires the use of a static magnetic field to polarize the nuclear spins, it would be insightful to study the effect of this perturbation on the dynamics of the $S = 1/2$ Heisenberg chain studied in this work. We believe that it would induce another crossover from superdiffusion to ballistic dynamics, which needs to be characterized.

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