

Entanglement in the pseudogap regime of cuprate superconductors

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We find a strongly enhanced entanglement within the pseudogap regime of the Hubbard model. This entanglement is estimated from the quantum Fisher information and, avoiding the ill-conditioned analytical continuation, the quantum variance. Both are lower bounds for the actual entanglement and can be calculated from the (antiferromagnetic) susceptibility, obtained here with the dynamical vertex approximation. Our results qualitatively agree with experimental neutron scattering experiments for various cuprates. Theory predicts a $\ln(1/T)$ divergence of the entanglement for low temperatures T , which is however cut off by the onset of superconductivity.

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Introduction. Measuring entanglement in macroscopic solid-state systems is extremely challenging as many particles participate and are measured simultaneously. Only very recently, first entanglement witnesses such as the one- and two-tangle [1,2] and the quantum Fisher information (QFI) [2–5] have been determined in antiferromagnetic spin chains [1–3], triangular-lattice quantum spin liquids [4], heavy fermion systems [5], and—more closely related to our work—cuprates in the strange metal regime [6]. For a review, see Ref. [7]. In this context, the QFI is particularly handy because it connects entanglement depth [8,9] to the susceptibility [10], making it accessible in neutron scattering experiments. For general reviews and further (typically experimentally not accessible) entanglement measures in many-body systems, we refer the reader to Refs. [11,12].

The QFI is given by the following integral over the imaginary part of the susceptibility $\chi(\omega)$ [10],

$$F_Q = \frac{4}{\pi} \int_0^\infty d\omega \tanh\left(\frac{\omega}{2T}\right) \text{Im}[\chi(\omega)], \quad (1)$$

with a temperature (T) cutoff for the lowest frequencies ω . If $F_Q > m$, the system is at least $(m+1)$ -partite entangled [10]. That means, if measuring one particle (or say its spin), m other particles (degrees of freedom) are modified through the collapse of the wave function. For neutron scattering experiments, it is challenging to properly subtract the background and to include the large- ω tail of the magnetic susceptibility when evaluating Eq. (1). On the theory side, the reliance on real frequencies poses a significant obstacle for many numerical many-body methods where it necessitates an ill-conditioned analytical continuation.

The temperature cutoff of Eq. (1) reflects the quantum nature of entanglement. The “classical” $\omega = 0$ susceptibility that signals ordering but not entanglement must not be included in F_Q [13]. This $\omega = 0$ component even diverges at a finite-temperature phase transition irrespectively of whether we have a classical or quantum model, and thus does not necessarily reflect any entanglement. Quantum correlations can remain short ranged while the correlation length and the $\omega = 0$ susceptibility diverge [14]. This is the reason why quantum critical systems and quantum spin liquids were hitherto at the focus of QFI research [1–5,10].

As for theory, entanglement in many-body solid state systems has traditionally focused on spin systems. Only recently also strongly correlated electron systems such as the Hubbard model received more attention [15–30]. Different numerical methods have been employed for studying different entanglement measures and witnesses, but research is still in its infancy. A more accurate modeling of cuprate superconductors requires more than the single band of the Hubbard model, which is quite obvious from the fact that holes generated by chemical doping go into the oxygen orbitals [31,32] with the copper orbitals remaining close to half filling. Nonetheless, the Hubbard model is the most commonly studied model for cuprates [33], as an effective model and because it describes a single Fermi surface of mixed oxygen and copper character as observed in experiment.

In this Letter, we report that the pseudogap (PG) regime of the two-dimensional Hubbard model shows a significant entanglement as measured by the QFI and the quantum variance (QV) [13,34] which we calculate directly from imaginary (Matsubara) frequencies $i\omega_n$ as [see Eq. (S3) of the Supplemental Material [35]]

$$I_Q = 8T \sum_{n=1}^{\infty} \chi(i\omega_n) \leq F_Q. \quad (2)$$

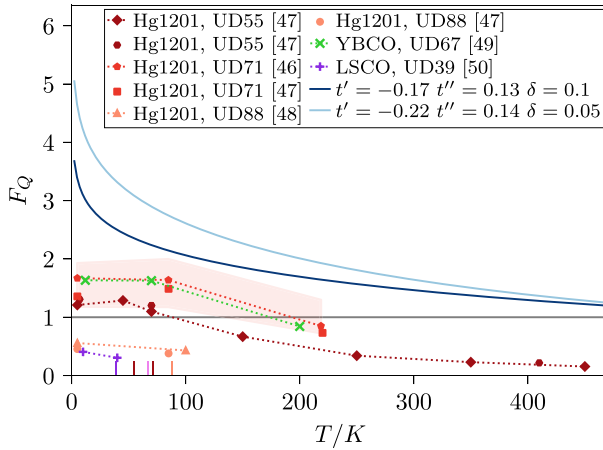


FIG. 1. Entanglement as witnessed by the QFI, comparing the Hubbard model at two different dopings δ and hopping parameters (solid lines; $t = -0.45$ eV and $U = 8t$ for both) to that calculated from various neutron scattering experiments [36]. The experimental data include large uncertainties, exemplarily indicated by the red shaded range for Hg1201 UD71. Importantly, the experimental QFI only includes the inelastic contribution, not the quasielastic contribution which is relevant in theory even with the QFI constraint $\omega > T$. Small vertical lines indicate superconducting critical temperatures (T_c) of each material, and the gray horizontal line indicates the onset of bipartite entanglement.

The QV I_Q is a lower bound to the QFI F_Q so that $I_Q > m$ also implies $F_Q > m$ and at least $(m + 1)$ -partite entanglement. Applying an Ornstein-Zernike fit, we also obtain the QFI and show that the entanglement of the pseudogap regime grows as $\ln(1/T)$. Our results for the Hubbard model compare favorably to neutron scattering experiments for various cuprate superconductors within the pseudogap regime (see Fig. 1, the main result of our Letter). One reason for the larger theoretical QFI is that the $\ln(1/T)$ divergence is cut off by the onset of superconductivity. Further, (i) the contribution from the quasielastic peak, important in theory, is missing in neutron scattering experiments due to the opening of a spin gap around T_c . (ii) To remove the background, the spectrum at a larger temperature is subtracted. These deviations appear to be more relevant than the uncertainties of the experimental error bars, which we highlight for HgBa₂CuO_{4+x} (Hg1201) at an underdoping corresponding to $T_c = 71$ K (UD71) (Hg1201 UD71) as the red shaded area. Moreover, the cutoff of the frequency integral leads to an underestimation of the value. We expect the deviation to be small, based on the theoretical observation of the summation error [35]. In any case, theory and experiment together demonstrate an enhanced entanglement within the pseudogap regime of cuprate superconductors.

Model and method. For investigating entanglement in the PG regime of cuprates theoretically, we consider the two-dimensional Hubbard model as a minimal model. Its Hamiltonian reads

$$H = \sum_{ij,\sigma} t_{ij} c_{i,\sigma}^\dagger c_{j,\sigma} + U \sum_i n_{i,\uparrow} n_{i,\downarrow}. \quad (3)$$

Here, $c_{i,\sigma}^\dagger$ and $c_{i,\sigma}$ are the creation and annihilation operators for an electron on lattice site i with spin σ ; $n_{i,\sigma} = c_{i,\sigma}^\dagger c_{i,\sigma}$ is the occupation number operator. We limit the hopping t_{ij} to nearest, next-nearest, and next-next-nearest-neighbor hoppings t , t' , and t'' , respectively; U is the on-site Coulomb repulsion. We use two sets of hopping parameters, $t' = -0.17t$, $t'' = 0.13t$ and $t' = -0.22t$, $t'' = 0.14t$, that cover a typical spread among different cuprates including HgBa₂CuO₄ (Hg1201) [37], but keep $t = -0.45$ eV fixed as it merely sets the overall energy scale. We vary the hole doping δ (measured relative to half filling) and U .

To solve the Hubbard model, we use dynamical mean-field theory (DMFT) [38] as implemented in w2DYNAMICS [39] as a first step and subsequently include nonlocal correlations beyond DMFT by the dynamical vertex approximation (DΓA) in its ladder variant with a λ correction [40–42]. This faithfully reduces the overestimated DMFT susceptibilities [41] to values that well agree with other numerical approaches [43]. Within DΓA, the local but dynamical DMFT vertex is used to build nonlocal generalized susceptibilities through the Bethe-Salpeter equation. For the Hubbard model, momentum-dependent and frequency-dependent spin fluctuations are dominating here. Subsequently, via the Schwinger-Dyson equation, a momentum-dependent self-energy is constructed from these generalized susceptibilities or the corresponding full vertex [41]. Thereby, in the two-dimensional Hubbard model spin fluctuations lead to the opening of the PG [41,44].

For the dominant antiferromagnetic (AFM) wave vector $\mathbf{Q} = (\pi \pm \Delta, \pi)$ or $(\pi, \pi \pm \Delta)$, the DΓA magnetic susceptibility $\chi(i\omega_n)$ at bosonic Matsubara frequencies $\omega_n = 2\pi Tn$ is used to compute the QV directly using Eq. (2). It has been shown that the susceptibility at a specific wave vector can be used to witness entanglement [45]. For our choice of the local operator, $O = \sum_j e^{iq_j} S_j$, maximal entanglement is witnessed at $\mathbf{q} = \mathbf{Q}$. In the Supplemental Material [35], we show how the incommensurability Δ depends on the doping δ . In the main part we only show $\Delta = 0$ results [except of the La_{2-x}Sr_xCuO₄ (LSCO) experiment]. Note that Eq. (2) remains a proper lower bound for entanglement, even if the summation is only performed up to a finite n ; and we apply a cutoff at half the bandwidth, $\omega_n < W/2 = 4(t + t'')$ if not stated otherwise. For calculating also the QFI, we fit $\chi(i\omega_n)$ to the Ornstein-Zernike (OZ) form

$$\chi(i\omega_n) = \frac{A}{\xi^{-2} + \gamma|i\omega_n|}, \quad (4)$$

and integrate Eq. (1) with the analytical continuation thereof and cutoff $W/2$. The temperature dependence of the amplitude A , correlation length ξ , and (boson) damping factor γ as fitted to the DΓA is shown in the Supplemental Material [35].

For the experimental data we digitized the magnetic susceptibility from the literature [46–50] with the background already subtracted and frequency ranges starting from between 0 and 40 meV and ranging up to 20–190 meV. A simple integration with the trapezoidal rule has been employed. In the same way as for the theory results, we calculate the QFI for the leading AFM wave vector $\mathbf{Q}$. To this end, experimental susceptibilities are rescaled by the g factor $g = 2$, $\chi^{zz} = \frac{1}{g^2} \chi^{\text{exp}}$,

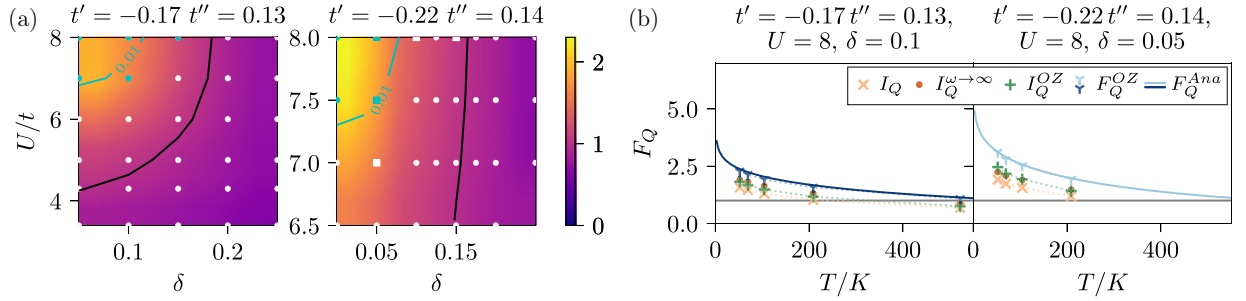


FIG. 2. (a) QV as a function of Coulomb repulsion U and doping δ for two sets of t', t'' and $T = 52$ K. Black lines indicate the region where at least a bipartite entanglement is detected, and (cyan/white) dots and squares represent computed data points (inside/outside) the PG regime; for the squares, the spectral function is shown in Fig. 3, and the cyan line displays a $\chi^{-1}(i\omega = 0) = 0.01$ contour that serves as a proxy for the onset of the PG [52]. (b) Comparison of QV from (i) a sum over Matsubara frequencies (I_Q), (ii) extrapolated to infinite frequencies ($I_Q^{\omega \rightarrow \infty}$), (iii) from the OZ fit (I_Q^{OZ}), and the QFI from (iv) the OZ fit (F_Q^{OZ}) and its analytical evaluation (F_Q^{Ana}). In the left figure, we display $t' = -0.17t, t'' = 0.13t$ at $U = 8t$ and $\delta = 0.1$ while in the right panel $t' = -0.22t, t'' = 0.14t$ at $U = 8t$ and $\delta = 0.05$ is shown. Dotted lines are guides for the eye and the horizontal gray line indicates the barrier for bipartite entanglement.

to ensure the correct normalization of the QFI [51]. Note that the subtraction of the background is very complicated in most experiments for both low and high frequencies, possibly leading to an underestimation of the QFI.

Even more importantly, we did not find properly normalized neutron data for the low-frequency quasielastic peak, which means that a relevant contribution to the QFI is missing. This certainly is a factor why the experimental QFI is smaller than the DGA calculated one. The latter to a large part indeed stems from the quasielastic peak [see Eq. (4)].

Results. In Fig. 2(a) we show the QV as a function of U and δ for the two sets of hopping parameters t', t'' considered. The QV drastically increases towards large U and small δ , within the PG regime. The cyan dots indicate those points for which we observe a PG in the spectral function (see Supplemental Material [35]). Here, within the PG regime the QV becomes at least three-partite for the studied temperature $T = 52$ K. Hence, in the PG regime, spins cannot be considered to be independent, and measurements performed on a single spin will strongly influence multiple other spins.

In the PG region, also the static susceptibility $\chi(\omega = 0)$ that does not contribute to the entanglement is large (cyan isovalue line). Indeed, the largeness of $\chi(\omega = 0)$ has been taken as a proxy for the PG [52], and empirically agrees very well with the opening of the PG [35]. Whether or not we have a pseudogap is shown exemplarily in Fig. 3 and Supplemental

Material [35]. Here,

$$-\frac{1}{\pi T} G(\mathbf{k}, \tau = 1/(2T)) = \frac{1}{2\pi T} \int d\omega \frac{A(\mathbf{k}, \omega)}{\cosh[\omega/(2T)]} \quad (5)$$

is an average of the spectral function $A(\mathbf{k}, \omega)$ over a frequency range $\pm T$ around the Fermi energy, that avoids the ill-conditioned analytical continuation.

Figure 2(b) presents the temperature dependence of the QV at fixed U and δ . In this figure, we also plot the QV summed over all Matsubara frequencies $I_Q^{\omega \rightarrow \infty}$. We see that this full integration actually only leads to a minor enhancement of I_Q compared to the summation up to the half-bandwidth cutoff. Further, we fit $\chi(\omega_n)$ to the OZ form, and integrate I_Q^{OZ} on the real frequency axis with the OZ fit according to Eq. (S7) of the Supplemental Material [35]. The good agreement between $I_Q^{\omega \rightarrow \infty}$ and I_Q^{OZ} validates the quality of the OZ fit [53]. Independent of the applied method, we observe that bipartite entanglement arises around room temperature or above, which is also the temperature range at which the PG first opens in cuprates [54].

Let us now turn to the QFI F_Q^{OZ} which is integrated on the real frequency axis with the OZ fit as well. As mathematically required, the QFI is higher than the QV, however only by about 1/2. Given the fact that the QFI by itself is only a rough lower bound of the actual entanglement this additional underestimation of the entanglement by 1/2 appears

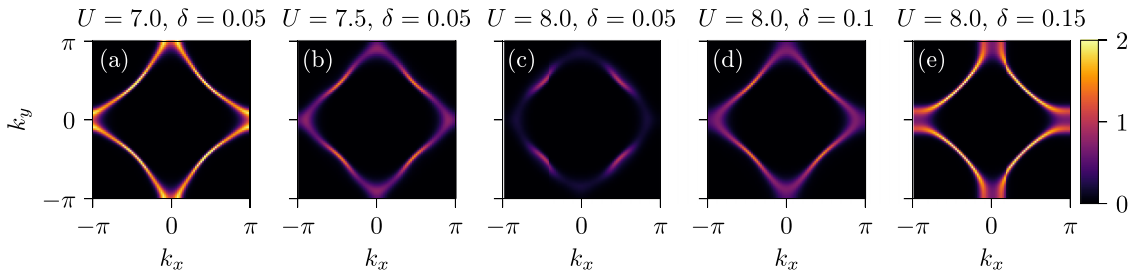


FIG. 3. Momentum-resolved spectral function around the Fermi energy $-\frac{\beta}{\pi} G(\mathbf{k}, \tau = \beta/2)$ showing the pseudogap opening with increasing U from (a) to (c) at fixed $\delta = 0.05$ and closing again at fixed $U = 8.0$ with increasing hole doping δ from (c) to (e). The panels correspond to the squares in Fig. 2(a) with $t' = -0.22t, t'' = 0.14t, U = 8t$, and $T = 52$ K.

bearable, in particular as the QV and QFI show the same overall trends.

Finally, we arrive in the Supplemental Material [35] at an analytic form of the QFI which holds in the limit of low T (and large correlation length ξ that is approached for low T):

$$F_Q \approx \frac{4}{\pi} \frac{A}{\gamma} \left[1 + \ln \left(\frac{W/2}{2T} \right) \right]. \quad (6)$$

This analytical form is plotted as a solid line in Fig. 2(b) and is in good agreement with the QFI from the OZ fit in the whole T range considered. It shows a $\ln(1/T)$ divergence for low temperatures. This divergence directly stems from the leading $1/\omega$ behavior for small ω in the OZ form Eq. (4), with temperature T acting as a cutoff. (There is another cutoff given by the inverse correlation length ξ^{-1} which is however smaller in two dimensions where ξ grows exponentially in $1/T$.) Note that the prefactor A/γ approaches a constant for $T \rightarrow 0$ (see Supplemental Material [35]). Physically, the term $\gamma\omega$ in Eq. (4) describes the overdamping of the bosonic susceptibility mode which is characteristic of strongly correlated electron systems. It describes the dynamical (quantum part of the) susceptibility in the frequency regime that is most relevant for the QFI estimate of entanglement. In contrast to the vicinity of quantum critical points, the QFI as a lower bound for entanglement is—to a first approximation— independent of the correlation length ξ within the PG regime. At a quantum critical point, the diverging correlation length would also lead to a diverging QFI with the same critical scaling [10].

In Fig. 1 we plot this analytical form of the QFI and compare it to neutron scattering susceptibility data extracted from various experiments for cuprate superconductors in the PG regime [46–50]. Both actually agree very well for HgBa₂CuO_{4+x} (Hg1201) at an underdoping corresponding to $T_c = 55$ K (UD55) and UD77, and to YBa₂Cu₃O_{6+x} (YBCO) UD67, demonstrating that in cuprates and the Hubbard model alike we have at least a bipartite entanglement as $F_Q > 1$ and, as discussed, experimental values are expected to underestimate the actual value. At least one reason why La_{2-x}Sr_xCuO₄ (LSCO) UD39 and Hg1201 UD88 have a lower QFI is that these are closer to optimal doping where the pseudogap gets weaker, and also in theory the QFI is considerably smaller.

An important difference from theory is that the experimental QFI saturates for low T . This saturation coincides with entering the superconducting phase (small vertical lines in Fig. 1), an aspect that is not captured by our theoretical calculation which does not allow for symmetry breaking.

For the bilayer cuprate YBCO, we show in Fig. 1 the QFI for $\mathbf{Q} = (\pi, \pi, 0)$ as we consider in our calculation only a single layer. We emphasize, that in this three-dimensional (3D) material at $\mathbf{Q} = (\pi, \pi, \pi)$ the QFI is twice as large, i.e., $F_Q \gtrsim 5$, which shows that there is not only entanglement within the layers but also between them [49].

Finally, we note that the quantum Fisher information (QFI) can also be computed from other normalizable response functions, such as density fluctuations. For the Hubbard model and parameters at hand, the QFI from the density-density susceptibility remains smaller than one, meaning that no entanglement is observed.

Discussion and conclusion. The Mermin-Wagner theorem [55] prevents AFM order for any finite temperature on an infinite and strictly two-dimensional lattice. There is however a $T = 0$ quantum critical line of AFM order up to a critical doping δ_c . In actual cuprates, finite sample sizes and even a weak hopping perpendicular to the CuO₂ planes lead to AFM order of the parent compounds [56]. Nonetheless, AFM is strongly suppressed and restricted to much smaller dopings than in a three-dimensional system. This leads to largely enhanced AFM fluctuations above the hypothetical quantum critical line of the ideal two-dimensional system. These enhanced AFM fluctuations are often considered to be the microscopic origin for the pseudogap and unconventional superconductivity in cuprates [57].

Here, we have shown that these strong antiferromagnetic fluctuations, with the $\omega = 0$ static contribution explicitly taken out, also mean enhanced entanglement within the pseudogap regime of the Hubbard model and in cuprate superconductors. This entanglement grows as $\ln(1/T)$ and only saturates when entering the superconducting phase. The multipartite entanglement was found for the antiferromagnetic spin susceptibility. We thus expect that when measuring one spin in the PG phase of cuprates, other spins (most likely neighboring ones) will be strongly affected through the collapse of the quantum mechanical wave function or density matrix.

The entanglement in the PG phase of the Hubbard model and in cuprates as deduced from the QFI and the QV, which is more generally accessible in theory, is comparable to that of other materials with strong entanglement such as quantum spin liquids and heavy fermion systems. As for the still lower experimental QFI in cuprates compared to theory, the ball is now in the court of neutron scattering experiment, which hitherto had a focus on the elastic peak. However, the quasielastic peak at $\omega > T$ is very important for the QFI.

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Data availability. The data that support the findings of this article are openly available [58].

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