Mott insulating states with competing orders in the triangular lattice Hubbard model

Alexander Wietek,1,* Riccardo Rossi,1,2 Fedor Šimkovic IV,3,4 Marcel Klett,5 Philipp Hansmann,6 Michel Ferrero,3,4 E. Miles Stoudenmire,1 Thomas Schäfer,5 and Antoine Georges4,1,3,7

1Center for Computational Quantum Physics, Flatiron Institute, 162 Fifth Avenue, New York, NY 10010, USA
2Institute of Physics, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland
3CPHT, CNRS, École Polytechnique, IP Paris, F-91128 Palaiseau, France
4Collège de France, 11 place Marcelin Berthelot, 75005 Paris, France
5Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany
6Department of Physics, University of Erlangen-Nürnberg, 91058, Erlangen, Germany
7DQMP, Université de Genève, 24 quai Ernest Ansermet, CH-1211 Genève, Suisse

(Dated: October 20, 2021)

The physics of the triangular lattice Hubbard model exhibits a rich phenomenology, ranging from a metal-insulator transition, intriguing thermodynamic behavior, and a putative spin liquid phase at intermediate coupling, ultimately becoming a magnetic insulator at strong coupling. In this multi-method study, we combine a finite-temperature tensor network method, minimally entangled thermal typical states (METTS), with two Green function-based methods, connected-determinant diagrammatic Monte Carlo (DiagMC) and cellular dynamical mean-field theory (CDMFT), to establish several aspects of this model. We elucidate the evolution from the metallic to the insulating regime from the complementary perspectives brought by these different methods. We compute the full thermodynamics of the model on a width-4 cylinder using METTS in the intermediate to strong coupling regime. We find that the insulating state hosts a large entropy at intermediate temperatures, which increases with the strength of the coupling. Correspondingly, and consistently with a thermodynamic Maxwell relation, the double occupancy has a minimum as a function of temperature which is the manifestation of the Pomeranchuk effect of increased localisation upon heating. The intermediate coupling regime is found to exhibit both pronounced chiral as well as stripy antiferromagnetic spin correlations. We propose a scenario in which time-reversal symmetry broken states compete with stripy-spin states at lowest temperatures.

I. INTRODUCTION

The interplay between strong electronic interactions and geometric frustration gives rise to a plethora of intriguing physical phenomena. It also raises fundamental questions that are still largely open, such as how insulating spin liquids transition into a metallic or superconducting phase upon reducing the interaction strength or introducing doped charge carriers.

Several classes of experimental platforms are available in which these questions can be explored. The most recent one is the rapidly developing field of twisted moiré heterostructures of two-dimensional materials, such as graphene [1–3] or transition-metal dichalcogenides [4, 5]. Recent work has demonstrated that these heterostructures provide a versatile platform for quantum materials design in which a broad range of lattice and band structures can be engineered [6, 7]. A triangular lattice structure, which is the focus of the present paper, can be realized in this context for B moiré superlattices [8, 9], twisted WSe2 double bilayers [10] as well as twisted bilayer Boron Nitride [11, 12]. The observation of a Mott insulating state in e.g. the WSe2/WS2 moiré superlattice system [8] provides direct experimental evidence of the importance of strong electronic correlations in these materials. We also note that the triangular superlattice dichalcogenide 1T-TaS2 has been proposed to host a spin-liquid state [13–16].

Besides moiré materials, strong electronic correlations in the context of (anisotropic) triangular lattice structures are also directly relevant to the two-dimensional molecular materials of the κ-ET family [17]. This class of materials has been the subject of intense experimental research and displays a diversity of remarkable phenomena (for reviews, see e.g. [18, 19]). Among those are Mott insulating phases with either magnetic long-range order or spin liquid behavior as in e.g. κ-(ET)2Cu2(CN)3, a pressure-induced metal-insulator transition (MIT). Several experiments found evidence of first-order phase transitions at finite-temperature up to a proposed critical temperature of ~ 20 K [20, 21]. Moreover, superconductivity with a critical temperature reaching ~ 14 K has been found. [21–31]. Finally, transition metal oxides such as the layered superconductor Li3NbO2 also form triangular lattices, with structural similarities to some of the dichalcogenides [32–34].

While the Hubbard model [35–38] on the triangular lattice is directly relevant to this wide variety of materials, it is also a paradigmatic model of strongly correlated electrons subject to geometric frustration and has therefore been subject to intense computational and theoretical research [39, 40]. However, due to the high complexity of the problem, only a partial understanding of its physics has been reached. The model is defined by the Hamilton-
\[ \hat{H} = -t \sum_{(i,j), \sigma} \left( \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + \hat{c}_{j\sigma}^\dagger \hat{c}_{i\sigma} \right) + U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}, \]  

where \( \hat{c}_{i\sigma}^\dagger, \hat{c}_{i\sigma} \) denote the fermionic creation and annihilation operators on site \( i \) with fermion spin \( \sigma \), \( \hat{n}_{i\sigma} = \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} \), and \((i,j)\) denotes summation over nearest-neighbor bonds of the triangular lattice.

At half-filling \((\hat{n}_{i\uparrow} + \hat{n}_{i\downarrow}) = 1\) the model has a metallic phase for small \(U/t\), while it is an insulator with long-range magnetic order in the large \(U/t\) limit for \( T = 0\). At finite temperatures true long-range magnetic order is prohibited in two dimensions by the Mermin-Wagner theorem \[41, 42\]. It has been suggested early on that an intermediate insulating phase without magnetic long-range order exists between these two phases, at intermediate \(U/t\) \[43\][44\]. The existence of this intermediate phase has been corroborated by several different computational methods \[45–52\]. Recent density matrix renormalization group (DMRG) studies showed strong evidence that the intermediate phase ground state realizes a gapped chiral spin liquid (CSL) \[52–54\]. This elusive state of matter has been proposed in the late 1980s \[55, 56\] and in the last years has been found to be stabilized in several frustrated spin systems \[57–63\], including extended triangular lattice spin-1/2 Heisenberg models \[64, 65\].

The demonstration of the emergence of topological superconductivity upon hole-doping the triangular lattice CSL \[66\] (see also \[67\]) constitutes an exciting prospect for the physics of the moiré materials and organic superconductors. Other previous suggestions on the nature of the intermediate phase include a Gutzwiller projected Fermi sea \[68–70\] and other forms of gapless spin liquids \[71–73\]. However, the existence of the intermediate CSL phase is also challenged by earlier DMRG results \[74\] as well as recent variational Monte Carlo studies, suggesting the absence of a spin liquid phase close to the metal-insulator transition \[75, 76\]. Other recent DMRG results on width 3 cylinders have suggested a gapless spin liquid being realized in the intermediate phase \[77\].

Computational methods for studying quantum many-body systems rely on diverse concepts and usually involve approximations whose validity has to be subject to critical evaluation. In the context of the Hubbard model, direct comparisons and benchmark studies involving multiple methods have proved successful in establishing the physics in the strongly correlated \[78–80\] and intermediate coupling \[81\] regimes beyond the uncertainties associated with the limitations of one particular method (see also a recent study on the kagome lattice \[82\]). This multi-method approach is currently playing a crucial role in the field and accelerates further theoretical and computational developments.

In this article, we combine conceptually different methods to investigate the physics of the half-filled triangular lattice Hubbard model at finite temperature. For some physical observables, the results from these different methods can be directly compared, but each method also comes with physical observables that it can more naturally address. Such a ‘multi-method, multi-messenger’ approach \[81\] therefore allows us to investigate the physics of this complex model from different perspectives.

On the one hand, we employ the minimally entangled thermal typical state method (METTS) \[83, 84\] which is an extension of DMRG to finite temperature. On the other hand, we use two Green function based techniques, the diagrammatic Monte Carlo method (DiagMC \[85\]) in its connected determinant version \[86–90\], dynamical mean-field theory (DMFT \[91–94\]) and a cluster extension \[95\] thereof: cellular DMFT (CDMFT) \[96, 97\] in its center-focused formulation \[98\]. Such a ‘handshake’ between wave-function based and Green function based methods is a notable advance which opens new perspectives for the study of quantum many-body problems at finite temperature.

Each of these methods has strengths and limitations which we now briefly describe. As a matrix-product state technique METTS can be applied with high precision on cylindrical geometries of finite circumference, as demonstrated recently by some of the authors in the case of the hole-doped square lattice Hubbard model \[99\], where a detailed description of the implementation of the method can be found. In this manuscript we mostly focus on a particular cylindrical geometry of circumference 4, called theYC4 geometry shown in Fig. 7. Selected results on the XC4 and YC3 geometries (see e.g. \[52\]) are also presented for comparison. Since we demonstrate convergence of our results in the maximal bond dimension \(D_{\text{max}}\) (Appendix A), the main limitation is in the finite transverse size.

As the triangular-lattice Hubbard model is afflicted by the fermionic sign problem, we cannot use traditional Quantum Monte Carlo techniques \[100\]. Diagrammatic Monte Carlo can work directly in the thermodynamic limit and is therefore immune from the sign problem, while being numerically exact: it is possible to compute quantities with arbitrary precision given enough computational time, and the convergence can be checked by comparing results from different expansion orders. Reaching the strong-coupling regime can, however, be hindered by the increased difficulty of resumming the perturbative series beyond their radius of convergence \[87\], and many orders must be computed, which in itself can present computational challenges. In this work, using the recent computational advances of the connected-determinant version \[86\], we are able to compute up to 10 orders of the perturbative expansion at fixed density; this is achieved \[101\] by renormalizing the chemical potential in the spirit of \[89\]. Thanks to the high orders reached, we manage to get converged results with controlled errorbars at temperatures \(T/t = 0.1\) and up to \(U/t = 10\).

CDMFT also works directly in the thermodynamic limit for the lattice, but it retains only a finite number of real-space components of the self-energy organized...
according to spatial locality and approximated by their value on a (self-consistent) cluster of finite size $N_c$. The method is controlled in the sense that it converges to the exact solution in the limit $N_c \to \infty$, but in practice, this convergence can only be reached in specific parameter regimes. Here, CDMFT is used in a twofold way: (i) as an approximation with $N_c = 7$ restricted to the paramagnetic (PM) phase [CDMFT-7 (PM)] and (ii) as an approximation with $N_c = 4$ (CDMFT-4) allowing for magnetic ordering. Unless noted otherwise the label “CDMFT” denotes results from the first variant. In either case on-site and nearest-neighbor components of the self-energy are taken into account, besides all temporal (quantum) correlations already present in single-site DMFT.

This article is organized as follows. We discuss the transition from a metallic state at weak-coupling to an insulating state at strong coupling in Sec. II. There, we perform a critical comparison between our numerical methods and propose that, in the accessible range of temperatures, a smooth crossover between these states is found rather than a first-order phase transition. In Sec. III we discuss the locality of the electronic self-energy by comparing results from CDMFT and DiagMC. In Sec. IV we investigate the basic thermodynamic properties of the system and firmly establish an order-by-disorder effect, where increasing temperature decreases the double occupancy. We relate this effect to an increase in entropy upon increasing the interaction strength via a Maxwell relation. Sec. V discusses competing (magnetic) orders as a function of temperature and interaction strength. In particular, we investigate magnetic structure factors and the chiral susceptibility to propose a scenario where chiral and stripy antiferromagnetic spin correlations coexist at low temperatures. Finally, we summarize and discuss our findings in Sec. VI.

II. METAL-INSULATOR Crossover

We begin by investigating the evolution from a metallic state at weak coupling to a Mott insulating state at strong coupling. At high enough temperature, this is a crossover. Whether it remains a crossover down to lowest temperatures or whether a phase transition also exists at low but finite temperature is discussed at the end of this section.

In order to identify this crossover, we consider two complementary observables, which are accessible within the CDMFT and METTS frameworks respectively. The first one is the zero-frequency value of the local (on-site) electronic spectral function:

$$A_c(\omega = 0) = -\frac{1}{\pi} \text{Im} G_c(i\omega_n \rightarrow i0^+).$$  \hspace{1cm} (2)

This quantity is evaluated by considering the central site of the cluster within the center-focused CDMFT method (see App. D and [98]) — hence the subscript in $A_c$. The extrapolation to zero frequency is obtained from a fit of the Matsubara frequency Green’s function $G(i\omega_n)$. We have also calculated within CDMFT the local and nearest-neighbor components of the self-energy and can extract the low-frequency slope:

$$Z_c = \left[1 - \frac{\partial \Sigma_c}{\partial \omega} \bigg|_{\omega \rightarrow 0} \right]^{-1},$$  \hspace{1cm} (3)

which is also a good indicator of the MIT. The non-local components of the self-energy are found to be quite small for weak to intermediate $U/t$ (see Sec. III for more details), hence $Z_c$ is a reasonable approximation in this regime to the spectral weight of quasiparticles. $A_c(0)$ is plotted in Fig. 1(a) as a function of $U/t$ for several values of temperature, along with $Z_c$ at $T/t = 0.1$ (inset). We see that for each value of $T$, $A_c(0)$ undergoes a marked drop as $U/t$ is increased, signalling a crossover from a metal with a large value of the zero-frequency spectral density to an insulator with a small one (but as expected still finite at non-zero $T$). Being a crossover there is a certain arbitrariness in defining its location precisely but it is apparent that, at the lowest temperatures, it occurs for

\[ \text{FIG. 1. Metal-Insulator crossover at finite-temperature from CDMFT and METTS. (a) Spectral weight at the Fermi level from CDMFT. The inset shows the quasiparticle renormalization factor for the central site in CDMFT (squares) as well as for PM-restricted DMFT (dots). A drop of the spectral weight at the Fermi level is observed. (b) Normalized localization length as a function of temperature and system size as obtained from METTS on the YC4 cylinder. Simulations have been performed with maximal bond dimension $D_{\text{max}} = 4000$. The normalized localization length attains a finite value in the insulating regime and increases in the linear system size $L$ in the metallic regime, where higher temperatures decrease the localization length.} \]
$8 \lesssim U/t \lesssim 9$. Correspondingly, $Z_c$ drops rapidly as $U/t$ is increased. At still larger values of $U/t$, the CDMFT self-energy has the characteristic divergent low-frequency behavior of an insulator, see Fig. 17 in Appendix D. Importantly, we see that $A_c(0)$ increases upon cooling for $U/t \lesssim 9$, while it decreases upon cooling for $U/t \gtrsim 9$, which are the expected behaviors in a metallic and an insulating regime, respectively. We note that for small temperatures and large interactions the CDMFT calculation becomes increasingly difficult due to the fermionic sign problem.

The evolution from a metallic state to an insulating state can also be characterized by the localization of electrons [102, 103]. An appropriate measure of localization is given by the localization length $\lambda$, which on open boundary conditions as employed by METTS is defined as [104–106],

$$\lambda^2 = \frac{1}{N} \langle \langle X^2 \rangle - \langle X \rangle^2 \rangle .$$

(4)

Here, $X = \sum_i r_i n_i$ denotes the position operator, where $r_i$ denote the coordinates of the lattice, $n_i$ the local density operators, and $N$ the total number of sites.

At zero temperature, the localization length $\lambda$ is directly related to the real part of the conductivity $\sigma(\omega)$ by [107]

$$\lambda^2 = \frac{\hbar}{\pi e^2 n} \int_0^\infty \frac{d\omega}{\omega} \text{Re} \sigma(\omega), \quad (T = 0)$$

(5)

where $e$ denotes the electron charge and $n$ the average density. The integral on the right-hand side, also referred to as Souza-Wilkens-Martin integral [104, 106, 107], diverges in the metallic regime and attains a finite value in the insulating regime for $N \to \infty$. The behavior of $\lambda^2$ for temperatures $T/t = 0.025, 0.100, 0.300$ and YC4 cylinder lengths $L = 8, 16, 24$ computed by METTS is shown in Fig. 1(b). The metallic and insulating regimes can be coarsely distinguished by the behavior of $\lambda^2$. Whereas in the insulating regime, $\lambda^2$ is almost constant as a function of the cylinder length $L$ and temperature, it increases with $L$ in the metallic regime. We also observe that at higher temperature, such as $T/t = 0.300$ in Fig. 1(b), the localization length decreases, indicating increased localization of the system.

Furthermore, we study the behavior of the charge structure factor given by,

$$S_c(k) = \frac{1}{N} \sum_{t,m=1}^N e^{ik \cdot (r_t - r_m)} \langle \langle n_t - \langle n_t \rangle \rangle \langle n_m - \langle n_m \rangle \rangle \rangle ,$$

where $n_l$ denotes the local density at site $l$. When using METTS we are working in the canonical ensemble with zero global charge fluctuation, which implies $S_c(0) = 0$ at any temperature. The behavior of $S_c(k)$ around $k = 0$ is indicative of metallic or insulating behavior. While a metallic state at $T = 0$ is characterized by a linear charge dispersion [108–111],

$$S_c(k) \approx \alpha |k_x|,$$

(7)

an insulating state exhibits a quadratic dispersion,

$$S_c(k) \approx \beta k_x^2 .$$

(8)

The behavior of the charge structure factor on the $16 \times 4$ YC4 cylinder from METTS at various temperatures in Fig. 2. We compare between the metallic regime at $U/t = 6$, the density structure factor behaves as $S_c(k_x) \approx |k_x|$. In the insulating regime at $U/t = 10$ we observe $S_c(k_x) \approx \beta k_x^2$. (c) Optimal fit parameters $\alpha, \beta$ for the ansatz $S_c(k_x) = \alpha |k_x| + \beta k_x^2$ of $S_c(k_x)$ close to $k_x = 0$. (resp. $\beta$) is shown as triangles (resp. circles). The crossover interaction strength $U_c/t$ can be defined by the intersection of $\alpha$ and $\beta$. We observe $U_c/t$ shifting towards weaker interactions at higher temperatures.

![Fig. 2. Static charge structure factor $S_c(k)$ for $k_y = 0$ at various temperatures (see legend in (c)) on the $16 \times 4$ YC4 cylinder from METTS with maximal bond dimension $D_{max} = 4000$. (a) In the metallic regime at $U/t = 6$, the density structure factor behaves as $S_c(k_x) \approx \alpha |k_x|$. (b) In the insulating regime at $U/t = 10$ we observe $S_c(k_x) \approx \beta k_x^2$. (c) Optimal fit parameters $\alpha, \beta$ for the ansatz $S_c(k_x) = \alpha |k_x| + \beta k_x^2$ of $S_c(k_x)$ close to $k_x = 0$. $\alpha$ (resp. $\beta$) is shown as triangles (resp. circles). The crossover interaction strength $U_c/t$ can be defined by the intersection of $\alpha$ and $\beta$. We observe $U_c/t$ shifting towards weaker interactions at higher temperatures.](image-url)
decreases the metallic weight $\alpha$ while increasing the insulating weight. In the insulating regime, we observe only a weak temperature dependence of $\alpha$ and $\beta$. We can define a crossover interaction strength $U_c/t$ by the intersection of $\alpha(U)$ and $\beta(U)$. We observe that $U_c/t$ shifts towards weaker interaction strengths when increasing temperatures. From this we estimate $U_c/t \approx 8.7$ at $T/t = 0.025$, $U_c/t \approx 8.5$ at $T/t = 0.100$, and $U_c/t \approx 8.0$ at $T/t = 0.500$. This also implies that, for a fixed $U/t$ in that range, the system undergoes increased localisation upon heating.

To further study the metal to insulator crossover we investigate the potential energy,

$$ E_{\text{pot}} = U \sum_i \langle \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} \rangle, \quad (10) $$

and the kinetic energy,

$$ E_{\text{kin}} = -t \sum_{(i,j),\sigma} \langle \hat{c}_{i\sigma} \hat{c}_{j\sigma} + \hat{c}_{j\sigma}^\dagger \hat{c}_{i\sigma} \rangle. \quad (11) $$

Since these quantities are accessible with all our methods, we show a direct comparison in Fig. 3(a,b) to assess the effects of finite cluster size in CDMFT and finite cylinder size in METTS. We focus on a temperature of $T/t = 0.1$, for which we perform numerically-exact simulations with DiagMC up to $U/t = 10.5$, which are used as benchmark. Remarkably, results from all methods agree within error bars up to an interaction strength of $U/t = 8$. Beyond this point, we still observe that the potential energy from METTS compares well with CDMFT [with $N_c = 7$ and restricted to its paramagnetic solution, CDMFT-7 (PM)] up to the strong coupling regime $U/t = 12$. A key difference between METTS and CDMFT is seen in the kinetic energy, which is lower for METTS than the data from single-site DMFT.

Above $T/t \approx 0.1$, all methods agree that the passage from the metal to the insulator is a smooth crossover [112]. Furthermore, it is clear from previous work [45–48, 52, 53, 113], that at $T = 0$ a metal-insulator phase transition (MIT) takes place. We now discuss whether our data allow us to settle whether a sharp MIT also exists at low but finite temperature or whether a smooth crossover applies for any non-zero temperature.

Let us first recall what the situation is in the single-site DMFT approximation. When solving the DMFT equations constrained to solutions without long-range magnetic order, one indeed finds that a first-order MIT develops for $T < T_c^{\text{DMFT}} \approx 0.1t$ ($U_c^{\text{DMFT}}/t \approx 11$) as previously demonstrated by several authors and also shown in Fig. 13 of Appendix C. Since there is no symmetry

![FIG. 3. Comparison of energies from different computational methods at $T/t = 0.1$. METTS results are obtained on a 16 $\times$ 4 YC4 cylinder with $D_{\text{max}} = 4000$, cluster DMFT is performed on a 7-site cluster [restricted to its paramagnetic solution, CDMFT-7 (PM)] and on a 4-site cluster (allowing for spin-symmetry breaking, CDMFT-4), while DiagMC is numerically-exact within the estimated errorbar. (a) Potential energy $E_{\text{pot}}$. METTS and cluster-DMFT agree with the DiagMC results up to $U/t \approx 7.5$. Beyond this point we find excellent agreements between cluster DMFT and METTS. (b) Kinetic energy $E_{\text{kin}}$. We observe good agreement between all methods up to $U/t = 8$. The inset shows the derivative of the kinetic energy w.r.t. the coupling strength $U$, which exists at $T/t = 0.025$ and $T/t = 0.100$. We observe a smooth dependence on the coupling strength and for both temperatures upon increasing the system size.](image-url)
as well as in cluster extensions of DMFT \[123–128\] when restricted to non-magnetic solutions. This transition is the reason for the cusp in the kinetic energy found with these methods, as apparent on Fig. 3(b) around \(U_{\text{CDMFT}}/t \approx 9\). However it should be emphasized that, when allowing for spin and translational symmetry breaking, the single-site DMFT approximation yields a solution with 120° Néel ordering for \(U/t \gtrsim 9.5\) [113] (see Appendix C): this is the true minimum of the free-energy in the DMFT approximation, hence overshadowing the first-order non-magnetic MIT. Incidentally, we note that the magnetically ordered DMFT solution yields a rather good approximation to both the kinetic and potential energy (Fig. 3).

Analogously to the DMFT case, we also computed the energetics within CDMFT on a \(N_c=4\) site cluster, however now allowing for magnetic symmetry breaking (CDMFT-4). The order sets in at similar interaction strengths as in the case of DMFT. Due to the smaller size of the cluster, and therefore the shorter non-local correlations that are included in the calculation, deviations from CDMFT-7 (PM) appear for the potential energy in the paramagnetic regime at intermediate coupling. At larger interaction strengths, when CDMFT-4 has ordered with a 120° Néel pattern, the potential energy acquires similar values to both METTS and CDMFT-7 (PM). In case of the kinetic energy, we observe deviations also in the ordered phase. They might root in the fact that the cluster geometries differ between CDMFT-7 (PM) and CDMFT-4 (for further details we refer to App. D). We note that calculations in the symmetry-broken phase for a \(N_c=7\) site cluster are not feasible at the moment due to the fermionic sign problem.

Of course, the magnetic solution at non-zero temperature is an artefact of the mean-field approximation inherent to DMFT, and one may argue that because fluctuations and Mermin-Wagner theorem actually prevent ordering, the existence of a finite-\(T\) MIT in paramagnetic DMFT/CDMFT is a hint that a similar phenomenon might take place in our model. On a qualitative level, frustration appears as a favorable factor by further suppressing ordering. The ET-organic materials with an (anisotropic) triangular structure do display such a transition experimentally [19].

Our METTS and DiagMC results do not provide evidence for such a first-order MIT or liquid-gas critical endpoint at finite temperature. In the range of temperatures that we could investigate, the kinetic and potential energy displayed in Fig. 3 do not appear to have a singularity as a function of \(U/t\). However, we acknowledge that limitations of our computational methods prevent us to reach a definitive conclusion about this issue. Our METTS results yield a smooth crossover between the metallic and the insulating regime for temperatures down to \(T/t = 0.025\). This is expected, since our simulations are performed on a finite system. Hence, observables will depend smoothly on the model parameters. We have, however, investigated the possibility of a discontinuity, indicative of a first-order phase transition, developing as a function of the length of the YC4 cylinder. Our results are shown for temperatures \(T/t = 0.025\) in Fig. 3(c) and for \(T/t = 0.100\) in Fig. 3(d). The smooth behavior at all system sizes does not exhibit any tendency to develop a discontinuity for \(L \rightarrow \infty\). However, it is possible that the chosen cluster geometry or the finite precision we achieve conceal potential singularities developing in the infinite-volume limit. The data points shown in Fig. 1 are spaced by \(\Delta U = 0.2t\) where the maximal absolute statistical error is of size \(\epsilon \approx 5 \cdot 10^{-3}\). The DiagMC results, while dealing with the infinite system, are limited in the present work to \(T/t \gtrsim 0.1\) and \(U/t \lesssim 10\). This is largely due to difficulties in computing enough expansion coefficients with small enough error bars to allow for controlled resummations of the perturbative series beyond the aforementioned values of \(U\).

### III. (NON-)LOCALITY OF CORRELATIONS: SELF-ENERGIES

Although the electronic Coulomb interaction is modelled as a purely local repulsion in the Hubbard model in Eq. (1), the correlations it generates can be non-local. To assess in which part of the phase diagram non-local
correlations become sizable in comparison to local ones, we calculate the local and nearest-neighbor (n.n.) self-energy in DiagMC and CDMFT in real space on the Matsubara axis. Fig. 4 displays the self-energy at the first Matsubara frequency $\Sigma(i\omega_0 = i\pi T)$ calculated by DiagMC (crosses) and CDMFT (squares) for two different temperatures (left panels: imaginary part, right panels: real part). The results of the calculations from both methods agree within error bars for both local and n.n. components.

At high $T/t = 0.40$ the correlations are mostly local and continuously increase from small to large $U$. However, there is an onset of non-locality already visible in the increase of the n.n. component at the largest interactions shown. The non-local correlations remain very small at lower $T/t = 0.10$ (close to the critical temperature of the MIT in CDMFT), until quite close to $U/t \approx 9.25$ at which the MIT takes place in CDMFT. Hence, through most of the metallic regime except close to the MIT, the self-energy in this temperature range is local to a good approximation in this frustrated system.

Upon entering the insulating regime, non-local correlations continuously increase. These non-local correlations signal increasing magnetic fluctuations. The onset of a magnetically ordered phase in DMFT (see App. C) underpins this interpretation. In the true solution of the system, of course, the Mermin-Wagner theorem [41, 42] prohibits magnetic ordering at finite temperature but the corresponding magnetic fluctuations are responsible for the increase of the (non-local) correlations. We note that the effect of magnetic fluctuations beyond DMFT using the dual fermion approximation has been investigated for this model in Refs. [47, 112, 129] and that the implications of non-local effects for transport has been investigated in Ref. [128].

IV. THERMODYNAMICS

We now turn to discussing the thermodynamic properties of the system for a range of interactions from $U/t = 6$ to $U/t = 12$. Figure 5 displays the specific heat $C$, thermal entropy $S$, internal energy $E$, and double occupancy $D$ as a function of temperature. Results for the specific heat,

$$C = \frac{\partial E}{\partial T},$$  \hspace{1cm} (12)

of the $16 \times 4$ YC4 cylinder using METTS at various values of $U/t$ are shown in Fig. 5(a). At intermediate and large interaction strengths $U/t = 9, 10, 12$ the specific heat exhibits a large mostly featureless plateau down to temperatures of $T/t \approx 0.1$. For $U/t = 10, 12$ a small peak develops at $T/t \approx 0.05$ before the specific heat tends towards zero at $T = 0$.

For $U/t = 6, 8$ we find a $T$-linear behavior of $C$ at low temperature, consistent with a metallic phase with gapless excitations. We note that the low-$T$ slope for $U/t = 8$ is approximately three times larger than that at $U/t = 6$: this is qualitatively consistent with $Z_e$ (inset of Fig. 1) being approximately three times smaller. For a metal in which the self-energy can be approximated as local, the quasiparticle effective mass enhancement which controls the slope of $C$ is related to the quasiparticle weight $Z$ by $m^*/m = 1/Z$. Indeed, as shown in the previous section, the non-local components of the self-energy are small through most of the metallic regime. Our findings for $Z_e$ and the slope of $C$ are thus consistent with quasiparticles developing a rather heavy mass as the insulator is approached. Although this is difficult to ascertain from our data, we find no evidence for a divergence of the effective mass when approaching the MIT however, consistent with the increasing non-locality of the self-energy in this regime (see Sec. III). We also observe that for $U/t = 6$, the specific heat appears to have another quasi-linear regime for $T/t \gtrsim 0.1$.

At larger interactions strengths the low-temperature behavior of $C$ also appears to be linear in $T$. However, given the few data points in this regime, it is difficult to
discern this behavior from other scenarios. We note that
the specific heat of the YC4 cylinder closely resembles the
specific heat that has been obtained on smaller clusters
using the finite-temperature Lanczos method [130].

We also investigate the thermodynamic entropy,
\[ S = \log(Z) + \frac{E}{T} = S_0 + \int_0^T d\Theta \frac{C(\Theta)}{\Theta}, \tag{13} \]
where \( Z \) denotes the partition function and \( E \) the inter-
nal energy. \( S_0 \) denotes a residual entropy at zero tem-
perature. The entropy is obtained by integrating the specific heat as in Eq. (13) from \( T = 0 \), where the internal (ground state) energy is computed by DMRG and we assume \( S_0 = 0 \), i.e. we assume a unique ground state on
the finite size cylinder.

Our results from METTS on the YC4 cylinder are shown in Fig. 5(b). Interestingly, we find that the en-
tropy (at fixed \( T \)) increases rapidly with increasing in-
teraction strength from the metallic regime at \( U/t = 6 \)
to the insulating regime beyond \( U/t = 9 \). The in-
crease in thermal entropy as a function of interaction strength has previously also been observed using the finite-temperature Lanczos method on smaller cluster geometries [130]. Naively, one would expect a decrease in entropy when the system is localizing. However, we find the exact opposite behavior, i.e. \( \partial S/\partial U > 0 \). This behavior is also reflected in the temperature dependence of the double occupancy,
\[ D = \frac{1}{N} \sum_{i=1}^N \langle n_{i\uparrow} n_{i\downarrow} \rangle, \tag{14} \]
shown in Fig. 5(d). The Maxwell relation
\[ \frac{\partial S}{\partial U} \bigg|_T = -\frac{\partial D}{\partial T} \bigg|_U \tag{15} \]
relates the increase in entropy as function of \( U \) to a de-
crease in double occupancy as a function of \( T \). Indeed, as shown on Fig. 5(d), we observe a decrease of \( D \) at low-\( T \) upon heating, for all displayed values of \( U/t \) up to a
temperature \( T/t \approx 0.5 \) at which the double occupancy has a minimum. On this figure, our METTS results are also compared to (C)DMFT which confirms our finding.

This phenomenon is analogous to the Pomeranchuk ef-
effect in liquid Helium 3 [131]. When the entropy of the insulating state is larger than that of the metal, increasing temperature indeed leads to increased localisation since this yields a gain in free-energy. To the best of our knowledge, this behavior was first predicted for the Hubbard model on the basis of DMFT studies [132] (see also [114, 115, 129, 133, 134]). It has been proposed [135] and experimentally realized [136] as a cooling scheme in the context of cold atomic gases in optical lattices. Inter-
estingly, it has also been recently observed in magic angle graphene [137]. In the present model, the large entropy of the insulating phase due to frustration and competing orders is responsible for this effect.

An important aspect of the thermodynamics is the be-
havior of the specific heat at low temperatures. In par-
ticular, the behavior of the specific heat allows us to dis-
tinguish between gapped and gapless phases. Here, we
investigate the internal energy \( E \) instead of the specific heat \( C = \partial E/\partial T \), since we measure the energy directly, whereas the specific heat is obtained from a numerical derivative of the energy. Our results for \( U/t = 8, 10, 12 \) are shown in Fig. 5(c). An analysis of the convergence as a function of bond dimension for \( U/t = 10 \) is shown in the appendix in Fig. 9. In the case of \( U/t = 8 \), we find that
the internal energy \( E \) approximately behaves as \( E \propto T^2 \)
for temperatures \( 0.02 \leq T/t \leq 0.1 \), which is indicated by
the black dashed lines. This translates to a \( T \)-linear be-
havior of the specific heat upon differentiation. Similarly,
for \( U/t = 10 \) and \( U/t = 12 \) the energy is well-described
by \( E \propto T^2 \) for behavior for \( 0.0125 \leq T/t \leq 0.4 \). We
notice, that the temperatures in this regime coincide with
region below the peak in the specific heat in Fig. 5(a).
For comparison, we also show a scaling \( E \propto T^{5/3} \) (i.e. \( C \propto T^{2/3} \)) as a gray dotted line, which is the
expected behavior of a spinon Fermi surface state [68–70]. As can be seen in Fig. 5(c) our data is in closer agreement to a \( E \propto T^2 \) than the \( E \propto T^{5/3} \) scenario. The \( T \)-linear behavior we observe would indicate a gapless state. However, we would like to point out that the lowest temperature attained in these simulations is \( T/t = 0.0125 \). Hence, our data does not rule out an activated behavior from a gap
smaller than this temperature. We would like to point out,
that these results are in agreement with experimental measurements of the specific heat for the triangular lattice compound \( \kappa-(ET)_2Cu_2(CN)_3 \) [138].

V. MAGNETISM

We study the magnetic properties as a function of tem-
perature and interaction strength of the system on the
YC4 cylinder using the METTS algorithm. Results on
magnetic ordering from (dynamical) mean-field theory
can be found in Appendix C. To distinguish different
kinds of orderings, we investigate the magnetic structure factor,
\[ S_m(k) = \frac{1}{N} \sum_{l,m=1}^N e^{i k \cdot (r_l-r_m)} \langle \hat{S}_l \cdot \hat{S}_m \rangle. \tag{16} \]
The momenta \( k \) resolved by the YC4 cylindrical geometry are shown in the inset of Fig. 6(a). Magnetic 120° Néel order can be detected by observing a peak in the structure factor at the K point in the Brillouin zone. On the YC4 cylinder, the K point is not exactly resolved, which is why we resort to the closest point \( K' \), shown in Fig. 6(a), to indicate 120° Néel order. A peak at the M-point can indicate the following two kinds of magnetic correlations:

(i) A collinear ‘stripy’ antiferromagnetic ordering is
characterized by breaking both spin and discrete \( C_6 \)
Several recent DMRG studies [52–54] have demonstrated that the triangular Hubbard model in the intermediate regime is susceptible to time-reversal symmetry breaking, which is indicated by a non-zero expectation value of the scalar chirality operator in the thermodynamic limit. To study such a scenario, we compute the chiral susceptibility

\begin{equation}
\chi = \frac{1}{N} \sum_{\mu,\nu \in \Delta} \langle \chi_\mu \chi_\nu \rangle ,
\end{equation}

where the scalar chirality operator on a triangle \( \mu = (l, m, n) \) is given by

\begin{equation}
\chi_\mu \equiv \vec{S}_l \cdot (\vec{S}_m \times \vec{S}_n). \quad (18)
\end{equation}

The sum in Eq. (17) extends over all pairs of elementary triangles. In the case of spontaneous time reversal breaking, we expect long range chiral correlations indicated by a large value of the chiral susceptibility \( \chi \). Since our simulations are working with real-valued wave functions, the expectation values of the scalar chirality operators, \( \langle \chi_\mu \rangle \), are exactly zero.

Moreover, we noticed a particular feature in our data on the YC4 cylinder, where prominent nearest-neighbor antiferromagnetic correlations switch between the different directions of the triangular lattice. To quantify this observation we introduce the nematic spin correlation,

\begin{equation}
\mathcal{N} = -\frac{1}{N} \sum_{\{i,j\}'} \langle \vec{S}_i \cdot \vec{S}_j \rangle ,
\end{equation}

where the sum extends only over nearest-neighbor pairs \( \{i,j\}' \) along the short direction of the cylinder (i.e. the direction pointing “north-east” in Fig. 7). The behavior of the above quantities as both a function of \( U/t \) and temperature \( T/t \) is shown in Fig. 6. We observe three distinct regimes as a function of \( U/t \). For \( U/t \lesssim 8.5 \) we do not observe any dominant magnetic features. As we have previously found in Fig. 1 and Fig. 2, this region corresponds to the metallic regime.

The intermediate regime ranging from \( 8.5 \lesssim U/t \lesssim 10.5 \) exhibits interesting and peculiar behavior in all observables. With decreasing temperature, magnetic correlations grow as signaled by the structure factor at both the \( K' \) and \( M \) points Fig. 6(b,f). But below the temperature \( T/t = 0.1 \) for \( U/t = 10 \), the \( K' \)-point structure factor begins to decrease, while at the \( M \)-point it increases sharply beginning around \( T/t = 0.05 \). The chiral correlations in Fig. 6(d) also increase below this scale, and the specific heat simultaneously develops a small maximum at \( T = 0 \) the system spontaneously breaks time-reversal symmetry and forms a chiral spin liquids. However, we also observe that the chiral correlations similarly build up beyond \( U/t \gtrsim 10.5 \).

Note that we use here the term ‘stripy’ in relation to spin degrees of freedom - we find no indication of a charge stripe density modulation.

(ii) Non-coplanar tetrahedral order, on the other hand, is formed when spins in a \( 2 \times 2 \) unit cell align in a way that they point towards the corners of a regular tetrahedron [64, 139].

FIG. 6. Magnetic ordering as a function of interaction strength \( U/t \) (left) and temperature \( T/t \) (right) on the \( 16 \times 4 \) YC4 cylinder from METTS. Simulations have been performed with a maximal bond dimension \( D_{\text{max}} = 3000 \). (a,b) Magnetic structure factor \( S_M(K') \) indicating \( 120^\circ \) Néel order. The inset in (a) shows the momenta resolved by the YC4 cylinder, and the position of the ordering vectors \( K' \) and \( M \) (c,d) chiral susceptibility \( \chi \) as defined in Eq. (17). Chiral correlations build up in both the intermediate as well as the strongly coupled regime at lower temperature. (e,f) Magnetic structure factor \( S_M(M) \) indicative of collinear stripy antiferromagnetic order. (g,h) Nematic spin correlation \( \mathcal{N} \), as defined in Eq. (19), is pronounced only in the intermediate coupling regime.
FIG. 7. Snapshots of METTS states $|\psi_i\rangle$ at temperature $T/t = 0.0125$ on the $16 \times 4$ YC4 cylinder in the intermediate regime at $U/t = 10$ (a) and in the strong coupling regime at $U/t = 12$ (b). The length of the arrows is proportional to the spin correlation $\langle \vec{S}_0 \cdot \vec{S}_i \rangle$, where the black cross marks the reference site. Hexagons and arrows which are blue indicate positive and red indicate negative spin correlations. The color of the triangles indicates the magnitude of the chiral correlation $\langle \chi_0 \chi_\mu \rangle$, where the reference triangle is indicated in gray just below the reference site. Nearest-neighbor spin correlations $\langle \vec{S}_i \cdot \vec{S}_j \rangle$ are indicated as the width and color of the bonds. We observe collinear stripy spin correlations at this temperature in the intermediate regime at $U/t = 10$.

As already pointed out before, in principle a peak at the M point in the intermediate regime could also indicate non-coplanar tetrahedral magnetic order [139]. However, by inspecting real space spin correlations we clearly observe the formation of stripy antiferromagnetism. Using METTS we can investigate “snapshots” of the system at a given temperature [99]. Briefly summarized, the METTS method decomposes the thermal density matrix into a sum over rank-1 density matrices corresponding to pure states [83, 84],

$$\frac{1}{Z} e^{-\beta H} = \sum_i p_i |\psi_i\rangle \langle \psi_i| ,$$

where $p_i \geq 0$ are real non-negative probabilities, $|\psi_i\rangle$ are the so-called METTS wave functions, and $Z$ denotes the partition function. The pure states $|\psi_i\rangle$ are sampled with probability $p_i$. We show the properties of a typical METTS wave function sampled in our simulations at $T/t = 0.0125$ and $U/t = 10$ in Fig. 7(a). The stripy spin correlations are clearly pronounced for this METTS state. We also observe sizeable chiral correlations $\langle \chi_0 \chi_\mu \rangle$ which are indicated by the color of the inner triangles. When comparing the snapshot at $U/t = 10$ in Fig. 7(a) to the snapshot at a larger $U/t = 12$ in Fig. 7(b) we observe that the nearest-neighbor spin correlations are more strongly pronounced along the short direction of the cylinder at $U/t = 10$, whereas for $U/t = 12$ the spin correlations on the other two directions are enhanced.

This motivates the definition of the nematic spin correlation $\mathcal{N}$ in Eq. (19). In Fig. 6(g,h) we observe that $\mathcal{N}$ is clearly pronounced only in the intermediate regime. We would like to point out that the increased magnetic correlations align with the stripy spin patterns, as can be seen in Fig. 7(a).

One last notable aspect of the intermediate regime we find is that increasing the temperature from $T = 0$ not only suppresses double occupancy, as shown previously in Fig. 5, but also increases the $120^\circ$ Néel order correlations. Therefore, the effect of increasing temperature is similar to the effect of further increasing the coupling strength $U/t$, which also both localizes the system and favors $120^\circ$ Néel order for $U/t \geq 10.5$.

The strong coupling regime $U/t \gtrsim 10.5$ is most prominently characterized by the increase in the magnetic structure factor $S_m(K')$ at lower temperatures, shown in Fig. 6(a,b). This is indicative of $120^\circ$ Néel order in the ground state. We observe strong antiferromagnetic correlations for $U/t = 12$ setting in at a temperature below $T/t = 0.05$, which again coincides with the small maximum in the specific heat observed in Fig. 5(a). We find that the behavior of the chiral correlations is similar to the intermediate coupling regime. In particular, $\chi'$ as shown in Fig. 5(d) is rather comparable between $U/t = 10$ and $U/t = 12$. 
VI. DISCUSSION

The physics of the triangular lattice Hubbard model at half-filling is coarsely organized in three different regimes as a function of the coupling strength, $U/t$: a metallic regime is followed by an intriguing insulating regime at intermediate coupling regime whose nature is currently hotly debated. At large interaction strength the system enters a magnetic insulating regime, where coplanar 120° Néel order is stabilized in the ground state. Evidence for the existence of an intermediate non-magnetic insulating regime is ample in the literature [43, 45–52, 54, 129] and clearly confirmed by several of our findings using multiple numerical methods.

We firmly establish the order-by-disorder effect at intermediate coupling $U/t$, where increasing temperature paradoxically leads to increased localization, as apparent in the double occupancy shown in Fig. 5(d). As discussed in Sec. IV, this effect is similar to the Pomeranchuk effect [131] observed when liquid Helium 3 solidifies upon heating, and previously found to occur for the Hubbard model in DMFT studies [114, 115, 132, 133, 135]. The decrease in double occupancy upon heating is confirmed by both our METTS and cluster DMFT results, where we found good quantitative agreement between these two very different numerical techniques. This observation suggests that localized excitations carry a large thermal entropy at low temperatures. This is consistent with the Maxwell relation Eq. (15) relating decreasing double occupancy to an increasing entropy with increasing temperature as well as increasing $S$ of $\Delta S_0$.

We have shown in Fig. 6, both the chiral correlations as well as the magnetic structure factor at the M point developed a maximum towards the M point. This is consistent with spontaneous time-reversal and parity symmetry breaking, as expected for a ground-state chiral spin liquid (CSL) [52–54], the peak in $S_m(M)$ is not necessarily related to the formation of a CSL. While a peak at the M point could in principle be indicative of non-coplanar tetrahedral order [130], we find that in the present geometry this peak is related to the formation of nematic, stripy antiferromagnetic correlations. This finding is backed up by a recent ground state DMRG study [54], which analogously found a peak in $S_m(M)$ in the intermediate regime, although the authors found these correlations to be only short-ranged at $T = 0$.

The occurrence of stripy spin correlations is remarkable given that most known instances of chiral spin liquids are stabilized in close proximity to non-coplanar magnetically ordered states [57, 59–61, 64, 65]. The melting of non-coplanar magnetic ordering has been suggested as a guiding principle to understand the formation of CSLs [140]. This seems to be rather different in the present case, where nematic collinear correlations and chiral correlations both develop as the temperature is decreased. In this context, the variational study of the triangular lattice Heisenberg model with an additional ring-exchange term performed in Ref. [71] is particularly interesting. The model with ring exchange can be thought of as an approximate low-energy effective Hamiltonian for the intermediate coupling regime [51, 68]. The authors compared variational energies of several Gutzwiller projected ansatz wave functions, including an ansatz for a gapped chiral spin liquid and a gapless nematic spin liquid, breaking rotational symmetry. While both of these wave functions have been shown to have a comparable, competitive energy, the gapless nematic state had the lower variational energy for this particular model. A more recent variational Monte Carlo study has similarly suggested the stabilization of a gapless nematic spin liquid in the context of the half-filled triangular Hubbard model, albeit upon adding further second nearest-neighbor interaction [75]. Remarkably, our finite-temperature METTS simulations now reveal exactly this competition between chiral and stripy spin correlations at finite, but low temperatures. While the recent DMRG studies [52, 54] provided strong evidence, that ultimately at $T = 0$ a CSL is formed on the investigated geometries, we now propose that stripy spin correlations become relevant immediately at finite temperatures. We would like to point out, that an estimate for the gap of the CSL has been stated in Ref. [52] by computing the domain wall tension to be $\Delta \approx 0.0065t$, which is below the lowest temperature $T/t = 0.0125$ we have been able to simulate using METTS on the $16 \times 4$ cylinder. It is worth pointing out that the effect of finite-temperatures on the system is non-trivial, as can be seen by the increase in $S_m(M')$ when increasing temperatures in Fig. 6(a), or the decrease of the double occupancy in Fig. 5(d).

This raises the question about whether a (rotationally-
symmetric) perturbation could stabilize nematic stripy (quasi-)order at $T = 0$. In particular, it would be interesting to find out whether indeed a nematic gapless spin liquid with algebraic spin correlations can be realized and to study its transition to the CSL. However, such a state will likely have larger quantum entanglement than the gapped CSL, rendering accurate DMRG computations more difficult.

We expect the balance between chiral or nematic spin correlations to be strongly dependent on the finite size geometry. As the DMRG studies on the YC3-6 and XC4 cylinders have shown [52–54], the precise nature of the ground state still has rather strong dependence on the exact shape of the cylindrical geometry. Therefore, a detailed comparison of our finite-temperature METTS data for different geometries is therefore highly desirable.

We have performed an in-depth comparison between different geometries in appendix B. There, we discuss results on YC4 cylinders of varying length and also results on the YC3 and XC4 cylinders [52]. We find that our results only weakly depend on the length of the YC4 cylinders. The specific heat of the YC4 cylinder closely resembles the specific heat obtained on the YC3 and XC4 cylinders. In particular, we observe that the maxima for different values of $U/t$ develop at comparable temperatures across all different geometries. Similarly, the Pomeranchuk effect of decreasing double occupancy as a function of temperature is clearly observed on all geometries. The correlations at low temperatures in the intermediate coupling regime differ, however. While both the YC4 and YC3 geometries exhibit pronounced stripy antiferromagnetic correlations, this is not observed on the XC4 cylinder. Also, the chiral susceptibility only smoothly increases as a function of $U/t$ on the YC3 and XC4 cylinders. This is in contrast to the YC4 cylinder, where we observe the onset of chiral correlations at smaller $U/t$ than the onset of 120° magnetic correlations.

At this point, we would like to comment that finite-size effects are expected to become less severe at higher temperatures, since correlation lengths typically decrease. It remains to be seen down to which temperature scale the finite-size cylinders can fully capture the two-dimensional limit.

Let us now focus on the strong coupling regime at $U/t = 12$. In the limit $U/t \rightarrow \infty$ the effective spin degrees of freedom are described by the antiferromagnetic Heisenberg model, whose 120° Néel ordered ground state features spin-wave excitations. However, besides spin-wave excitations several authors have found a different kind of excitation being relevant in this case. Series expansions found anomalous behavior of the magnon spectrum, which exhibits a minimum beyond the description of linear spin-wave theory [141–143]. This minimum is ascribed to the presence of a different kind of excitations, reminiscent of the roton excitations of $^4$He forming a minimum in the quasiparticle dispersion [144, 145]. Therefore, the excitations of the triangular lattice Heisenberg antiferromagnet have often been referred to in literature as “rotonlike” excitations (RLE). It has been argued that these excitations contribute significantly to the thermal entropy down to temperatures $T \approx 0.1J$ [142], where $J$ denotes the antiferromagnetic coupling constant. More thoroughly, the presence of two different kinds of excitations in the $S = 1/2$ triangular Heisenberg antiferromagnet has been recently using the exponential tensor renormalization group (XTRG) method [146, 147], which similar to METTS, allows for unbiased numerical simulations at finite-temperature on cylindrical geometries. The authors indeed establish two temperature scales corresponding to the magnon and the RLEs. The RLEs are shown to manifest themselves in an increase of the nearest-neighbor chiral correlations as well as a maximum in the magnetic structure factor $S_{\alpha\beta}(M)$ [147]. The existence of the RLEs is demonstrated to be robust on a wide variety of cylinder geometries, including cylinders of circumference $W = 6$.

In our simulations at $U/t = 12$ we analogously find a maximum in $S_{\alpha\beta}(M)$ in Fig. 6(e) indicating the RLEs. Also an increase of chiral correlations in Fig. 6(d) at temperatures below $T/t \approx 0.1$ is observed. The 120° magnetic correlations at lower temperatures are signalled by a peak in $S_{\alpha\beta}(K')$. This clearly resembles the situation encountered in the Heisenberg model as in Ref. [147]. Interestingly, we found the specific heat in Fig. 5 to be rather similar for both the intermediate and strong coupling regime. It appears that tuning the interaction strength from $U/t = 10$ to $U/t = 12$ interchanges the role of stripy correlations with 120° correlations.

We would like to elaborate on the behavior of the chiral susceptibility $\chi$. If time-reversal symmetry is indeed broken at low-temperature in the intermediate regime, we would expect the chiral correlations to diverge towards $T = 0$. However, we think that the temperatures ($T \geq 0.0125t$) we simulated are above the transition temperature estimated by Ref. [52], $\Delta \approx 0.0065t$. Furthermore, it has also been argued that the stabilization of a CSL at $T = 0$ can depend on the cylinder length studied in DMRG [54], where a CSL at $T = 0$ has only been found in cylinders of length $L = 64$, but not in shorter cylinders. Nevertheless, we find pronounced chiral correlations in the intermediate coupling regime. For the strong coupling regime, pronounced chiral correlations at finite-temperature have been found in a previous study of the Heisenberg model using XTRG [147]. There, the correlations have been attributed to the rotonlike excitations of the triangular lattice Heisenberg antiferromagnet. Here, we find that the chiral correlations already build up in the intermediate regime and remain sizeable in the strong coupling regime. This strongly suggests that the rotonlike excitations are relevant excitations in the intermediate coupling regime.

Finally, an outstanding question is the occurrence of superconductivity in the present model. While general arguments suggest that the metallic phase studied here hosts a low-temperature superconducting instability at weak coupling (see e.g. [148]), the possible occurrence
of an unconventional superconducting phase near the metal-insulator phase boundary [45] or upon doping the insulating phase [67] are intriguing questions for future computational studies.

**ACKNOWLEDGEMENTS**

We are indebted to Nils Wentzell, Steven White, Michael Zaletel, and Sabine Andergassen for insightful discussions and support. We thank Elio König for valuable comments on the manuscript. We thank the computer service facility of the MPI-FKF, and the Scientific Computing Core of the Flatiron Institute for their help. METTS results were obtained using the ITensor Library (C++ version) [149] and CDMFT computations used the TRIQS library [150]. This work was granted access to the HPC resources of TGCC and IDRIS under the allocations A0090510609 attributed by GENCI (Grand Equipement National de Calcul Intensif). The authors gratefully acknowledge use of the computational resources of the Max Planck Computing and Data Facility. The present work was supported by the Austrian Science Fund (FWF) through the Erwin-Schrödinger Fellowship J 4266 - “Superconductivity in the vicinity of Mott insulators” (SuMo, T.S.). It has also been supported by the Simons Foundation within the Many Electron Collaboration framework. A.G. also acknowledges the support of the European Research Council (ERC-2013-AdG - Project QMAC-319286). The Flatiron Institute is a division of the Simons Foundation.

**APPENDIX**

**Appendix A: Convergence of METTS simulations**

We employ the METTS algorithm as described in Ref. [99]. Thermal expectation values of an operator $O$ are evaluated as,

$$\langle O \rangle = \langle \psi_i | O | \psi_i \rangle,$$

where the minimally-entangled typical thermal states,

$$| \psi_i \rangle = e^{-\beta H/2} | \sigma_i \rangle,$$

are imaginary-time evolved product states $| \sigma_i \rangle$. Here, $\sigma_i$ denotes statistical averaging over a series of subsequent METTS. As such, the METTS algorithm is subject to statistical sampling uncertainty, which can be reduced by computing more samples and whose size can be estimated using standard time series analysis. The imaginary-time evolution is performed by using the time-dependent variational principle (TDVP) algorithm for matrix product states [151–153]. In Ref. [99], some of us showed that the maximal bond dimension $d$ of the matrix product state representation of the METTS serves as a control parameter to achieve accurate and controlled computations on finite size cylinders. Here, we performed extensive comparisons between simulations at different bond dimensions. Results from simulations performed with $D_{\text{max}} = 1000, 2000, 3000, 4000$. Results agree for all quantities within errorbars already at $D_{\text{max}} = 1000$. Comparisons are performed at $T/t = 0.025$ (left) and $T/t = 0.1$ (right). We show the double occupancy $D$ (a,b), the magnetic structure factors evaluated at $M$ (c,d) and $K'$ (e,f), and the chiral susceptibility $\chi$ (g,h).

**FIG. 8.** Convergence of METTS results on the 16 × 4 YC4 cylinder as a function of maximal bond dimension $D_{\text{max}}$. We compare results from simulations performed with $D_{\text{max}} = 1000, 2000, 3000, 4000$. Results agree for all quantities within errorbars for $D_{\text{max}} = 2000, 3000, 4000$, whereas results at $D_{\text{max}} = 1000$ deviate slightly. Comparisons are performed at $T/t = 0.025$ (left) and $T/t = 0.1$ (right). We show the double occupancy $D$ (a,b), the magnetic structure factors evaluated at $M$ (c,d) and $K'$ (e,f), and the chiral susceptibility $\chi$ (g,h).
FIG. 9. The internal energy $E$ as a function of $T/t$ at $U/t = 10$ on the $16 \times 4$ YC4 cylinder depending on the maximal bond dimension $D_{\text{max}}$. We compare results from simulations performed with $D_{\text{max}} = 2000, 3000, 4000$ and find our results to agree within errorbars. The behavior of the energy is found to be well described by $E \propto T^2$, implying a $T$-linear specific heat $C$.

Appendix B: Comparison of METTS cylinder geometries

The results from METTS in the main text have mainly been obtained on the $16 \times 4$ YC4 cylindrical geometry, which is shown in Fig. 7. In this appendix, we discuss the effects of the cylinder geometry on our results. First, we investigate the dependence of the magnetic observables on the cylinder length $L$ of the YC4 cylinder in Fig. 10. We compare simulations at temperatures $T/t = 0.025, 0.100, 0.300$ for $6 \leq U/t \leq 12$. We find that our results only weakly depend on the cylinder length $L$ and are, therefore, expected to be robust in the limit $L \to \infty$ for the YC4 cylinder.

Moreover, we assess the effect of the cylinder width and boundary conditions on our results. Here, the differences between the geometries are more pronounced, and the physics at lowest temperatures in the intermediate coupling regime differs in some aspects. This has previously already been observed in Refs. [52, 53], where a detailed comparison of DMRG results on different geometries has been performed. We focus on the YC3 and XC4 geometries. The YC3 geometry is similar to the YC4 geometry shown in Fig. 7, but has a circumference of $L_y = 3$. The YC3 geometry allows for stabilizing the $120^\circ$ Néel order, and features both the K and M points in reciprocal space. The XC4 geometry, on the other hand, has a circumference of $L_y = 4$, but differs from the YC4 geometry by having a distinct periodicity vector given by $T = (0, \frac{\sqrt{12}}{2} L_x)$. The $120^\circ$ Néel order is unfrustrated on this lattice and both K and M points are featured in reciprocal space. The resolved momenta of the YC3 and XC4 geometry are shown in Fig. 11 and Fig. 12, respectively.

We show the behavior of the specific heat $C$, magnetic structure factor $S_m(q)$, double occupancy $D$, and chiral susceptibility $\chi$ for the YC3 cylinder in Fig. 11(a) and the XC4 cylinder in Fig. 12(a). First, the behavior of the specific heat is similar on all cylinder geometries we investigated. At $U/t = 6$ we observe a broad maximum at $T/t \approx 0.25 - 0.4$. This maximum is shifted towards lower temperatures of around $T/t \approx 0.15$ at $U/t = 8$. In the intermediate to strong coupling regime, we observe a small maximum at temperatures around $T/t = 0.05$ followed by an extended plateau. At low temperatures, we observe an increase of the specific heat as a function of $U/t$, which by Eq. (13) implies an increase in entropy upon increasing $U/t$. As discussed in Sec. IV, this increase in entropy is related to a decrease of the double occupancy $D$ with temperature via the Maxwell relation Eq. (15). The double occupancy $D$ for the YC3 and XC4 geometries is shown in Fig. 11(c) and Fig. 12(c). The decrease in double occupancy is clearly observed in all geometries. Hence, the Pomeranchuk effect is consistently realized on all geometries we investigated.

The magnetic structure factor $S_m(q)$ at temperature $T/t = 0.025$ as a function of $U/t$ is shown in Fig. 11(b) for the YC3 cylinder and Fig. 12(b) for the XC4 cylinder. The YC3 cylinder exhibits a clearly pronounced...
peak at \( \mathbf{q} = \mathbf{M} \) in the intermediate coupling regime. This is consistent with our results on the YC4 cylinder, where we similarly detected stripy antiferromagnetic order. Also, at strong coupling the structure factor is peaked at \( \mathbf{q} = \mathbf{K} \), indicating \( 120^\circ \) Néel order. However, we do not observe a pronounced chiral susceptibility \( \chi \) in the intermediate coupling regime in Fig. 11. Instead, the chiral susceptibility smoothly increases as a function of \( U/t \). This is consistent with the ground state DMRG study performed in Ref. [52], where no chiral spin liquid has been observed for periodic boundary conditions on the YC3 cylinder.

Similarly, the XC4 cylinder also does not exhibit a pronounced chiral susceptibility in the intermediate coupling regime in Fig. 12(d). Also for this geometry, Ref. [52] reported the chiral spin liquid not being realized at \( T = 0 \) for periodic boundary conditions. In contrast to the YC3 and YC4 geometries, the XC4 cylinder does not exhibit a pronounced peak of the magnetic structure factor at the \( \mathbf{M} \) point in the intermediate regime, as shown in Fig. 12. Instead, the peak at \( \mathbf{q} = \mathbf{K} \) is smoothly increasing as a function of \( U/t \). This demonstrates that the precise nature of the state realized in the intermediate regime at low-temperatures is dependent on the particular cylinder geometry. We think, the kind of order being exactly realized in the two full-dimensional limit is still to be determined. However, the pronounced the stripy antiferromagnetic correlations on the YC3 and YC4 cylinder and the evidence for a CSL on the YC4 cylinder [52, 54] show that these two kinds of orderings are competing at the lowest temperatures, and might be realized in the full two-dimensional limit, possibly upon adding further interaction terms.

Appendix C: Magnetic phase transition in (dynamical) mean-field theory

In this Appendix we give an overview of the magnetic properties of the Hubbard model on the isotropic triangular lattice calculated by means of the dynamical mean-field theory (DMFT). By the inclusion of all temporal and spatial correlations present in the Hubbard model Eq. (1), DMFT has proven to provide a good starting point for the application of more sophisticated techniques, which aim at including spatial correlations on top (see, e.g., [47, 81, 129, 154]).

The main panel of Fig. 13 shows the Néel temperature calculated in DMFT \( T_N^{\text{DMFT}} \) (red line and circles) and static mean-field theory (MFT, dashed line), separating the paramagnetic from a magnetically ordered phase [155]. In contrast to the case of the Hubbard model on a square lattice where due to the nesting properties of the Fermi surface the order appears for every finite \( U \) at low enough temperatures, here a quantum critical point separates a Fermi liquid from a magnetically ordered ground state at \( U_{\text{QCP}}^{\text{DMFT}}/t \approx 9.5 \) (cf. also [47, 129]).

From there \( T_N^{\text{DMFT}} \) increases steeply with a maximum of \( T_{N,max}^{\text{DMFT}}/t \approx 0.25 \) around \( U/t = 11 \) before slowly decreasing again.
Two points are particularly noteworthy:

(i) as in the case of the Hubbard model on a square lattice the critical end point of the Mott MIT (orange triangle) visible in the paramagnetically restricted DMFT is shadowed, i.e. preempted, by the magnetic phase transition of DMFT (with the magnetically order phase being the thermodynamically stable phase of DMFT) and

(ii) the CDMFT critical end point (blue square) lies close to the phase boundary of the magnetic phase. Please note that we did not calculate the magnetic phase diagram in case of CDMFT, which is restricted to its paramagnetic solution.

Comparing the magnetic phase diagram of DMFT to the self-energies of CDMFT at \( T/t = 0.10 \) presented in Fig. 4 of the main text, one can observe that the nearest-neighbor component of the CDMFT self-energy starts to increase at the interaction value \( U/t = 9.5 \), where the DMFT orders magnetically. In other words the spatial mean-field approximation reflects the increase of non-local fluctuations by entering an ordered phase.

For the determination of the DMFT phase boundary, we calculated the momentum-dependent magnetic susceptibility \( \chi_{\text{m}}^{\text{DMFT}}(k, \Omega_n = 0) \) at zero Matsubara frequency by means of the solution of the Bethe-Salpeter equations with the irreducible vertex extracted from the self-consistently determined Anderson impurity model [93], using the continuous time quantum Monte Carlo solver in its interaction expansion (CT-INT) and the tprf framework [150] of TRIQS [150]. For the vertex we used up to \( N_{\text{pw}} = 50 \) positive fermionic Matsubara frequencies and extrapolated the value of the physical susceptibility.
to \( N_i \omega \to \infty \) with \( \chi \sim a + b/N_i \omega \) (see, e.g., Supplemental Material of [157]).

Due to the second order nature of the phase transition, approaching the phase boundary \( \chi_{\text{DMFT}}(k, i\Omega_n = 0) \) diverges at the ordering vector \( k = Q \). Fig. 14 shows \( \chi_{\text{DMFT}}(k, i\Omega_n = 0) \) at \( T/t = 0.40 \) (left column) and \( T/t = 0.10 \) (right column) for several interaction values. One can see that the leading contribution always stems from momentum vectors centered around \( k = K \). Approaching the transition, at \( T/t = 0.10 \), \( \chi_{\text{DMFT}}(k, i\Omega_n = 0) \) continuously grows before it eventually diverges at \( k = K \).

The temperature dependence of the inverse susceptibility \( \chi_{\text{DMFT}}^{-1}(k, i\Omega_n = 0) \) is shown in Fig. 15 for two different values of the interaction. At \( U/t = 8 \) (with a Fermi liquid ground state present in DMFT) it exhibits Pauli-like behavior, i.e. approaching a constant at low temperatures. At \( U/t = 10 \) (with a magnetically ordered ground state) the susceptibility diverges as \( \chi \sim |T - T_N|^\gamma_T \) at \( T_R^{\text{DMFT}}/t \approx 0.22 \) with \( \gamma_T = 1 \) being the susceptibility’s mean-field critical exponent.

Appendix D: Cellular dynamical mean-field theory: cluster geometry, Matsubara data and computational details

a. CDMFT on a \( N_c = 7 \) site cluster, restricted to the PM solution [CDMFT, CDMFT-7 (PM)]

For the CDMFT calculations performed in this work we used \( N_c = 7 \) sites which are arranged according to Fig. 16 with a central site and six equivalent sites that form an outer ring. We restrict the CDMFT to its paramagnetic solution. Due to the previously found observation [98] that the self-energy obtained from a cluster center focused extrapolation converges faster with the cluster size than the periodization schemes previously introduced in the literature, for single-particle observables

\[
\begin{align*}
\mathbf{a}_1 &= \left(5/2, \sqrt{3}/2 \right) \\
\mathbf{a}_2 &= \left(1/2, 3\sqrt{3}/2 \right)
\end{align*}
\]

FIG. 16. Cluster geometry with \( N_c = 7 \) used in CDMFT. The cluster consists of a central site (marked in red) and six equivalent outer sites arranged on a ring. The translation vectors are \( \mathbf{a}_1 = \left(5/2, \sqrt{3}/2 \right) \) and \( \mathbf{a}_2 = \left(1/2, 3\sqrt{3}/2 \right) \).
central weight at low frequencies and (ii) a change of slope
\cite{158, 159}) and eventually divergence of the imaginary
part of the self-energy on the central site with increas-
ing $U/t$. The data of Fig. 1 (a) in the main have been
obtained from a linear extrapolation of the data in the
upper panel of Fig. 17 to zero frequency.

For the calculations of the self-energies, we converged
the CDMFT self-consistency cycle using a continuous
time quantum Monte Carlo solver in its interaction ex-
pansion (CT-INT) in the TRIQS framework \cite{150}.

b. CDMFT on a $N_c=4$ site cluster, allowing for
spin-symmetry breaking (CDMFT-4)

For the CDMFT calculations that allow for spin-
symmetry breaking (Fig. 3, CDMFT-4) we used a $N_c=4$
cluster with the geometry given in Fig. 18. As already
stated in the main text, the convergence of a larger clus-
ter (at least close to phase transitions) is prohibited by
the fermionic sign problem.

For being able to enter the symmetry-broken phase
we use an approach similar to the one described in \cite{113}
for single-site DMFT. However, instead of rotating the
Green function we rotate an external magnetic field $H\sigma_z$
by applying:

$$e^{i\phi\sigma_y/2}[H\sigma_z]e^{-i\phi\sigma_y/2}$$

where $\sigma_i, i \in \{x, y, z\}$ denote the Pauli matrices. The
120° Néel ordering corresponds to a rotation with $\phi = 2\pi/3$.

\[\mathbf{Q} = (2\pi/3, 2\pi/3)\]. The breaking of the $SU(2)$-
symmetry of the Green function results in an effective
8-orbital calculation at the impurity level. After some
iterations of the algorithm we switch off the field and let
the system converge self-consistently.

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