Tensor network investigation of the double layer Kagome compound Ca$_{10}$Cr$_7$O$_{28}$

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Quantum spin liquids are exotic quantum phases of matter that do not order even at zero temperature. While there are several toy models and simple Hamiltonians that could host a quantum spin liquid as their ground state, it is very rare to find actual, realistic materials that exhibit their properties. At the same time, the classical simulation of such instances of strongly correlated systems is intricate and reliable methods are scarce. In this work, we investigate the quantum magnet Ca$_{10}$Cr$_7$O$_{28}$ that was recently discovered to exhibit properties of a quantum spin liquid in inelastic neutron scattering experiments. This compound has a distorted bilayer Kagome lattice crystal structure consisting of Cr$^{7+}$ ions with spin-1/2 moments. Coincidentally, the lattice structure renders a tensor network algorithm in 2D applicable that can be seen as a new variant of a projected entangled simplex state algorithm in the thermodynamic limit. In this first numerical investigation of this material that takes into account genuine quantum correlations, good agreement with the experimental results is found. We argue that this is one of the very first studies of physical materials in the laboratory with tensor network methods, contributing to uplifting tensor networks from conceptual tools to methods to describe real two-dimensional quantum materials.

INTRODUCTION

Since Anderson’s proposal of the resonating valence bond state as one of the possible mechanisms of high $T_c$ superconductivity [1], the study of quantum spin liquids has attracted intense attention, both from theorists as well as experimentalists [2–8]. Such phases of matter, besides falling beyond the paradigm of Landau symmetry breaking theory, exhibit intriguing physical properties such as fractional statistics of their excitations or a degeneracy of the ground state manifold that depends on the topology of the problem, and feature no local order parameter. They are seen as key building blocks in topological quantum computing [9] where it is the absence of local order parameters that protects non-locally encoded quantum information against local noise and errors. While several theoretical models have been proposed that could host quantum spin liquids as ground states [7, 10, 11], finding actual materials natively exhibiting such properties which can be studied in a laboratory seem rare. This is mostly because realizing a quantum spin liquid in two spatial dimensions requires complex frustration mechanisms which often need to be fine tuned. For this reason, it is imperative to identify numerical simulation methods that can constructively guide such efforts.

Meanwhile, the rapid development of novel, advanced theoretical and numerical techniques for studying complex quantum many-body systems continues to provide strong impetus to understanding problems such as the one described above. While there are several different techniques available for this purpose, many of them do not explicitly take into account the most important ingredient in strongly correlated models in general and quantum spin liquids in particular: These are genuine quantum correlations in the ground state [12–14]. Even quantum Monte Carlo methods, among the most popular techniques known, fails to provide a tool here, due to the intrinsic sign problem commonly arising in such frustrated systems [15]. Tensor network (TN) techniques, in contrast, are built upon and constructed from notions of entanglement, exhibit neither of the limitations and provide a highly controlled set of powerful methods [16–19]. This design feature, thus, renders these techniques particularly well-suited to study frustrated systems and spin liquids. While such TN algorithms are ubiquitous and widely used in systems of one spatial dimension, their higher dimensional counterparts are significantly more challenging and complicated in implementation. However, the two-dimensional TN states known as projected entangled pair states (PEPS) [20, 21] have recently been maturating and can now be reliably used in numerical analysis of two-dimensional systems, providing state-of-the-art results when comparing all available methods known to problems at hand [22, 23].

In this work, we develop notions of projected entangled pair states to new instances of projected entangled simplex states (PESS) [24] that are applicable to capture the physics of a quantum magnet Ca$_{10}$Cr$_7$O$_{28}$ [25]. This is indeed a quantum material that recently was studied using several experimental techniques and was discovered to have properties of a quantum spin liquid [26, 27]. Heat capacity found no magnetic transitions down to temperatures of $T = 300$ mK and muon spectroscopy revealed the complete absence of any static magnetism even at 19 mK, but rather showed that the spins were moving coherently in the ground state below 0.4 K [26]. Inelastic neutron scattering revealed that the magnetic excitations are broad at all energies suggesting a multiparticle spectrum consistent with spinons which are the excitations of a quantum spin liquid [26]. The spectrum also appeared to be gap-less within the resolution implying that Ca$_{10}$Cr$_7$O$_{28}$ is a gap-less spin liquid. This work aims at bringing endeavours
of developing tensor network models for strongly correlated systems in two spatial dimensions to a new level, giving rise to one of the first instances of studying an actual material that can be prepared in the laboratory using sophisticated tensor-network based schemes – and not just a paradigmatic model. Thus, our results can be directly compared to experimental data and used as a benchmark in further investigations, providing precisely the type of guidance to experimental efforts hinted at above. In what follows, we will start by describing in detail our material Ca$_{10}$Cr$_7$O$_{28}$. We then turn to introducing the numerical technique that we employ to investigate this material. Equipped with this powerful tool, we will demonstrate that it exhibits features of a gap-less quantum spin liquid, a result concomitant with findings in the experiment. Complementing these results, we will compute the magnetization curve and the magnetic susceptibility of the model in the presence of an external magnetic field, again to good agreement with experiments. In an outlook, we present further perspectives of the approach taken.

**MATERIAL**

The material that we investigate is known as Ca$_{10}$Cr$_7$O$_{28}$. Powder samples of this material were prepared using solid state synthesis, while single crystals were grown using the travelling solvent floating zone technique. Details on how to prepare this compound in the lab can be found in Refs. [25, 28]. The crystal structure of this compound was determined using neutron diffraction experiments. It was found to consist of Cr$^{5+}$ ions which have spin-$1/2$ moments arranged in a bilayer breathing Kagome structure [25]. The two Kagome layers in this compound are different from each other and each of them again consists of two inequivalent alternating triangles (two different layers of breathing Kagome). The crystal structure of our compound is illustrated in Fig. 1.

The Hamiltonian of this compound consists of five inequivalent Heisenberg interactions that can be written as

$$H = (J_{\text{inter}} + J_{\Delta 1} + J_{\nabla 1} + J_{\Delta 2} + J_{\nabla 2}) \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j$$  \hspace{1cm} (1)$$

where the first term with interaction strength $J_{\text{inter}}$ corresponds to the coupling between the two layers in the compound and is ferromagnetic in nature. The second term with strength $J_{\Delta 1}$ corresponds to the coupling of the up triangles in the first layer and is also ferromagnetic in nature. $J_{\nabla 1}$ is for the antiferromagnetic down triangle in the first layer. Similarly, for the second layer, the up ($J_{\Delta 2}$) and the down ($J_{\nabla 2}$) triangles have anti ferromagnetic and ferromagnetic coupling, respectively. The nature and the precise strength of these coupling parameters were determined in Ref. [27] and are summarized in Tab. 1.

Later on, we will add an external magnetic field along the $S^z$ direction to obtain the magnetization curve of the material.

**TABLE I:** Values and nature of the Heisenberg interaction in the crystal structure of Ca$_{10}$Cr$_7$O$_{28}$ illustrated in Fig. 1. The interaction type ‘F’ and ‘AF’ corresponds to ferromagnetic and antiferromagnetic interaction.

| Exchange parameter | Coupling strength (in meV) | Interaction type |
|--------------------|----------------------------|------------------|
| $J_{\text{inter}}$ | -0.08(4)                   | F                |
| $J_{\Delta 1}$    | -0.27(3)                   | F                |
| $J_{\nabla 1}$    | 0.09(2)                    | AF               |
| $J_{\Delta 2}$    | 0.11(3)                    | AF               |
| $J_{\nabla 2}$    | -0.76(5)                   | F                |

The modified Hamiltonian then takes the form

$$H_{\text{field}} = H + \sum_i g_s \mu_B h S_i^z$$ \hspace{1cm} (2)$$

where $g_s$ is the g-factor whose value is approximately 2. $\mu_B$ is the Bohr magneton whose value is approximately $5.7883818012 \times 10^{-5}$ eV per Tesla and $h$ is the strength of the external field. We will obtain the ground state of the Hamiltonians in Eq. (1) and (2) using our PESS technique described in the next section.
NUMERICAL TECHNIQUES

Compared with other existing numerical techniques, TN algorithms are very well suited for the study of frustrated systems for reasons discussed in the previous sections. In fact, both one and two-dimensional TN algorithms have been applied in the past to study paradigmatic two-dimensional frustrated systems [29, 30]. In particular, two-dimensional TN algorithms have been extensively used in the past to study the Kagome Heisenberg antiferromagnet, the ground state of which has taken center stage in an elaborate academic debate on being either a $\mathbb{Z}_2$ gapped spin liquid [11, 31–35] or a $U(1)$ gapless spin liquid [30, 36–38]. The mineral Herbstmthite currently provides the best physical realisation of this model [39].

There is an intricate feature that this lattice has: The case of the Kagome lattice exhibits the presence of corner sharing triangles. There are two known possible ways to use two-dimensional TN algorithms for such lattices. The first possibility is to use conventional pair-wise entangled PEPS directly in the Kagome lattice. This can be done by either mapping the Kagome lattice onto a square lattice (at the expense of an overhead) or directly updating every two-sites in the Kagome lattice. For instance, mapping the spins in the Kagome to a square lattice as in Refs. [40, 41], we saw that the local tensors are of the order of $D^5$ and $D^4$. Directly updating two sites without mapping to a square lattice would require local tensors of the order of $D^4d$ and $D^2$. Here $D$ and $d$ correspond to the bond and physical dimension of the Tensor network. On top of this computational bottleneck, neither of these techniques capture the important multi-partite entanglement of the frustrated system in a natural fashion. The second possibility is to use so-called projected entangled simplex states (PESS), generalizing PEPS. This technique updates three sites in a unit triangle and therefore captures the multi-partite entanglement in a very natural fashion. There are advantages in computational effort as well: the local tensors involved are of the order of $D^3$ and $D^2$ [24]. Hence, the latter is more efficient. It is this seemingly innocent feature that allows to go to very large bond dimensions (which is also essential if we want to study the entanglement scaling). For these reasons, we will use the PESS technique in our study of this double layer Kagome compound. A detailed discussion comparing the two techniques can be found in Ref. [40]. To the best of our knowledge, while such PESS techniques have been used in the past to study paradigmatic models like the Kagome Heisenberg antiferromagnet [30, 42], its application to pragmatic and realistic quantum materials in the lab has never been explored before. In what follows, we will describe our PESS technique briefly and explain how it can be used to study the compound Ca$_{10}$Cr$_7$O$_{28}$ in the focus of our study, which can be synthesized and explored experimentally.

PESS (PEPS) algorithms involve basically two stages: (i) the update scheme that refines the tensors and (ii) the contraction scheme that allow to compute properties. The first scheme is when we update the tensors, for example, in this case do an imaginary time evolution to obtain the ground state. The contraction scheme is required because we aim at computing the expectation values once we have the ground state. Let us describe the schemes that we employ in this investigation below.

Update scheme

In our investigation, we use a 3-site 3 PESS and a 3-site 9 PESS [24]. What this means is that we will update three tensors in a triangle (which is the basic unit of frustration in this lattice) at once while doing the update. We can do this by taking three (or nine) unique physical tensors in the thermodynamic limit repeated everywhere. In our case, we map two spins from the two layers of the Kagome compound to a single site. This means that, effectively, we have a six site and an eighteen site unit cell. The physical tensors are connected to each other by what is called the simplex tensors. Thus, there are two simplex tensors corresponding to the up and the down triangle in the 3-site 3-PESS case and six in the 3-site 9-PESS (later referred to as the six and eighteen site unit cell, respectively). Thus, this kind of update captures the multi-partite entanglement in a more natural fashion compared to the pair-wise entangled PEPS. This is shown in Fig. 2 for the 3-site 3 PESS (six site unit cell).

We employ a simple update scheme because of its numerical efficiency. We will compute the full environment when we calculate the expectation values. In order to make the update, we start by decomposing our Hamiltonian into two parts which do not necessarily commute i.e. $H = H_\Delta + H_C$. The first part of the Hamiltonian acts only on the tensors making the up triangle while the second part acts on the down trian-
The algorithm proceeds by applying $e^{-\delta \tau H_{\Delta}}$ and $e^{-\delta \tau H_{V}}$ successively on some random initial PESS state vector (distributed according to uniformly distributed random numbers in the interval $(0, 1)$). This means that the normalized state vector $|\psi\rangle = e^{-\tau H}|\psi_0\rangle$ in the limit of $\tau \to \infty$ is expected to provide the ground state vector upon convergence, where $|\psi_0\rangle$ is some initial state captured as a PESS [24]. The same applies for the case of the 9-site (eighteen site) unit cell where we apply the projection operator to six different triangles successively corresponding to nine physical tensors to obtain the ground state vector. We use successive Trotter steps of $10^{-1}$, $10^{-2}$, $10^{-3}$ and $10^{-4}$. Thus the error coming from the Trotterization is at most of $O(\delta \tau^2)$ i.e. $O(10^{-8})$. Thus, once we reach convergence, the ground state vector in the PESS formalism can be expressed mathematically for the 3-site 3-PESS as

$$|\psi\rangle = \text{Tr}(X R_{\mu}^{a,d,b',c'} A^{a',i,a,b} B^{b',i,b,c} C^{c',i,c} |\cdots, i_a, i_b, i_c, \cdots\rangle)$$

(3)

where $R_{\mu} = R_{\Delta}$ or $R_{V}$, the simplex tensors of dimensions $D \times D \times D$ with $D$, the bond dimension of the PESS, and $X$ denotes the entire rest of the contracted tensor network. It connects the tensors $A$, $B$ and $C$ associated with physical sites, each having physical dimension $d^2$ and bond dimension $D$, so that the entire tensors are of degree three and dimension $D \times D \times d^2$. It is key to the approach taken here that $d^2$ is the physical dimension of the double layer system of spin-1/2 systems, here seen as one physical system of local dimension four. The above expression is for the 3-site 3-PESS, one can express similarly for the 3-site 9-PESS.

**Contraction scheme**

Once we have obtained the ground state vector in the form of PESS, we need to be able compute expectation values of the Hamiltonian as well as of observables. This can be done by either using a simple environment or a full environment of the tensors. We will do both. In the case of the simple environment, we neglect correlations beyond a certain cluster and therefore do not need to contract the whole TN in the thermodynamic limit. There are numerical evidences suggesting that such approximations give very accurate results [43]. For the full environment, we need to contract the TN with a bond dimension that is the square of the original bond dimension. In both worst case and average case complexity, this is known to be a computationally hard problem for exponential precision [44, 45]. This statement can, however, be lifted for ground states of uniformly gapped systems. Indeed, in practice, there are several excellent algorithms that can efficiently and accurately approximate the full environment. Corner Transfer Matrix Renormalization Group (CTMRG) [46, 47], boundary Matrix Product States (bMPS) [21], and channel environment [48] are some examples. A detailed work comparing the different contraction schemes will be presented in the future. In this work, we will use the CTMRG technique to compute the environment. We describe it briefly below.

First of all, we regroup the physical and simplex tensors in the original Kagome lattice so that we obtain a square lattice which can be contracted in a straightforward manner using the CTMRG technique. Obviously, such a grouping is not unique. We choose the grouping as shown in Figure 3. The TN shown there corresponds to the overlap of the state vector $|\psi\rangle$ and the bra $\langle \psi |$ vectors. This means that for a PESS with bond dimension $D$ description of the wavefunction, we actually need to contract a PEPS with bond dimension $D^2$ to compute the energy or the observable. Figure 3 shows the 3-site 3-PESS (six site unit cell for our double layer compound). The resulting square lattice from this grouping has a two-site unit cell as shown in Figure 3(b). Once the square lattice is ready, we approximate the tensors in the shaded region in Fig. 3 using four corner matrices and a number of half-row and half-column transfer matrices depending on the number of unit cells. Details on how to compute these fixed point tensors can be found in Ref. [46, 47]. While computing the environment for a particular site, we fix the bond dimension of the CTM tensors to some number $D_{\text{CTM}}$. This is known as the bond dimension of the environment. The physical dimension of the CTM tensors is given by the bond dimension $D$ of the PESS quantum state. In our case, this value is $D^2$ since we need to compute the overlap of the vectors and dual vectors while calculating the expectation values.

**RESULTS**

Having described the numerical techniques used in this paper briefly, we will now present the results obtained using them. We will compute the ground state energy as well as the magnetization in different directions as a function of the
bond dimension of the PESS. We will then apply an external magnetic field and obtain the magnetization curve. The results will then be discussed and compared with the experimentally obtained ones.

Ground state energy

To start with, we will compute the ground state energy of our Hamiltonian, i.e., $\langle \psi_0 | H | \psi_0 \rangle / \langle \psi_0 | \psi_0 \rangle$ for $| \psi_0 \rangle$ being a good approximation of the ground state vector obtained using our update scheme. We will compute this quantity for different values of the bond dimension $D$ of the PESS. Importantly, we will compute the expectation value using two approaches: (i) simple environment (SE): that takes into account exact tensor contraction only within a certain cluster and (ii) full environment (FE): that takes into account the full contraction of the tensors in the thermodynamic limit. Intuitively, the simple environment is expected to be a very good approximation for systems with a short correlation length as it ignores correlation beyond a cluster. For this reason, we do not have to do the full contraction of the PESS tensors and hence we are able to go to a remarkably large bond dimension: $D = 20$ in our case. The results are shown in Fig. 4. From the plots, we can observe an algebraic convergence of the ground state energy as a function of the bond dimension of the PESS $D$. It is clear that the scaling does not depend on the number of unit cells used in our computation (six and eighteen). Such a behaviour is indicative of the fact that our model is critical or gap-less in nature [49]. Similar conclusion has been drawn on such scaling for the KHAF model, also using PESS techniques [30].

The calculations for the ground state energy in Fig. 4 were done using simple environment. To show that our results are not an artefact of the simple environment used, we have plotted the energy as a function of the bond dimension of the PESS using the full environment. We use the CTMRG algorithm for this purpose with bond dimension $D_{\text{CTM}}$. The results are shown in Fig. 5. For each $D$ of the PESS shown in Fig. 5, we made sure that the energies are well converged with the bond dimension of the environment $D_{\text{CTM}}$. For instance, for $D = 6$, the ground state energy is converged up to the 5th decimal place with increasing bond dimension of the environment $D_{\text{CTM}}$. For $D = 8$, the energies are converged up to 6th decimal places with the bond dimension of the environment. The results are similarly well converged up to significant number of digits for other bond dimensions of the PESS $D$ except for $D > 11$. For $D > 11$, it becomes increasingly difficult to calculate the full environment computationally for relatively high bond dimension of the environment. It is worth pointing out here while $D$ corresponds to the PESS bond dimension of the ground state, in practice, we are actually contracting a PEPS network with bond dimension $D^2$. For example, to compute the full environment of $D = 20$ PESS, we actually need to contract a 2D TN with bond dimension 400 which is computationally very difficult at the moment. For this reason, we compute the full environment for very large $Ds$ only.
up to the best possible value of $D_{\text{CTM}}$, that is computationally feasible to us as the moment. Interestingly, we find that the numbers obtained using the simple environment are sufficiently accurate. To quote some numbers we find that for $D = 6$ and $D = 8$, the simple environment is accurate up to 4th decimal places (not shown). For this reason, we strongly believe that the scaling behaviour of the plot in Fig. 4 will remain unaffected in the presence of the full environment. This is also supported by the plot in Fig. 5. In Ref. [30] in the study of the KHAF, the authors move to large bond dimensions (up to $D = 25$) and use “full environment”. We would like to point out that their technique is based on treating physical indices as bond indices. Such a technique then use the existing PEPS contraction scheme to contract the new TN. However, one should be aware of the fact that such a transformation of physical index into bond index may lead to unpredictable results, as one is also effectively truncating the physical dimension of the Hilbert space.

**Magnetization**

Having computed the ground state energy as a function of the bond dimension $D$ of the PESS using both the simple and the full environment, we will now compute the net magnetization per site of the ground state vector for the parameters in Tab. I. This is key to our analysis, as this can be directly measured in the experiment. Our results are shown in Fig. 7.

In Fig. 7, we show the average magnetization per site $M = (M_x^2 + M_y^2 + M_z^2)^{1/2}$ for different bond dimensions $D$ of the PESS ground state vector, where $M_x$, $M_y$, and $M_z$ correspond to the expectation value of the Pauli operators. The plot above is for the six site unit cell, while the one below is for eighteen site unit cells. We would like to mention again that our results do not depend on the number of unit cells used in our calculations. This is also indicative of a quantum spin liquid preserving the lattice symmetry. Both calculations have been done using the simple environment. We can see that the net magnetization disappears even for small bond dimensions of the PESS. The absence of symmetry breaking along any directions provides a strong evidence for a SU(2)-symmetric quantum spin liquid for the ground state. The algebraic convergence of the energy in Fig. 4 and this absence of magnetic ordering strongly suggests that the ground state is indeed a gap-less spin liquid. Our results are consistent with experimental findings of the same compound as revealed by
Mean field approximations in a ‘product-like’ state and in the presence of strong magnetic field, the ground state is maintained by mean field results [27]. This is to be expected since, our theoretically obtained curve as the temperature gets lowered by our tensor network ansatz for the specific choice of parameters working with a one-to-one comparison can be made with the experimental data [26, 27]. In the previous subsections, we have investigated the properties of the compound Ca_{10}Cr_7O_{28}, defined by the Hamiltonian \( (1) \) for the parameter regimes in Table I. We would now turn to investigating the response of this material to the presence of external magnetic field. To be precise, we would like to find the magnetization curve of the Hamiltonian in Fig. (2) in the presence of a strong external field. Our results are displayed in Fig. 8. The plots are obtained using a eighteen site unit cell calculation for \( D = 1, 2 \) and 3. Our result can be compared with the experimental data [27]. We observe a linear curve, without the existence of any magnetization plateaux as in the case of KHAF. The linear curve saturates at around \( 1 T \). The experimental magnetization curve also shows a steep and approximately linear increase in magnetization up to a field of \( H = 1 T \), however, it only achieves \( 50\% \) of the saturation magnetization at this field and continues to increase with a much gentler slope up to saturation at \( 12 T \) (see Ref. [26], fig. 2b). This discrepancy might be due to the fact that the experimentally obtained magnetization curve was measured at \( T = 1.8 K \) and one can expect the behaviour to converge to our theoretically obtained curve as the temperature gets lowered. Our results are also very similar to theoretically obtained mean field results [27]. This is to be expected since, in the presence of strong magnetic field, the ground state is in a ‘product-like’ state and mean field approximations will be very accurate. In fact, the mean field results can be reproduced by our tensor network ansatz for the specific choice of \( D = 1 \). This is also shown by the blue curve in Figure 8.

In addition to the magnetization curve, we have computed the magnetic susceptibility \( \chi \), which is nothing but the slope of the magnetization curve \( (dM/dh) \). We obtain a value of \( \chi = 1.1731 \mu_B/(Cr^{5+} \cdot T) \) at low values of the magnetic field \( (0 - 0.2 T) \). This is shown in the inset of Fig. 8. In other units, this value of susceptibility corresponds to 3.93 emu/(mol*Oe). This is very close to the experimentally obtained value of \( \chi \) at \( T = 1.8 K \) which has been found to be 3 emu/(mol*Oe). This agreement is hence not only on the purely qualitative, but also on a quantitative level.

**CONCLUSION**

In this work, we have set out to investigate the properties of the quantum magnet \( \text{Ca}_{10}\text{Cr}_{7}\text{O}_{28} \) with tensor network methods that are particularly suitable for studying such quantum materials due to the favourable scaling in effort. We found strong evidence of a gap-less spin liquid from the algebraic convergence of the ground state energy and the disappearance of magnetic order parameter using finite entanglement scaling. We have also investigated the response of this material to applying an external magnetic field, and have computed the magnetization curve as well as the magnetic susceptibility. Our theoretical results agree well with the available experimental results including the value of susceptibility measured in experiment at \( T = 1.8 K \).

Our studies constitutes the first instance of using state of the art tensor network tools to benchmark the properties of actual two-dimensional quantum materials that can be designed and studied in the laboratory. In this sense, our study marks a paradigm shift in the treatment of TN tools which were previously used mostly for paradigmatic theoretical models, albeit providing significant conceptual insight. The highly encouraging results found here provide a road map for future endeavours of benchmarking quantum materials with tensor networks. We plan to make our study even more realistic by studying the finite temperature properties of this model so that a one-to-one comparison can be made with the experimental data. Initial steps taken by us in this direction in developing annealing algorithms for 2D tensor networks [50] suggest that this can be done. It is the hope that the present work firmly places tensor networks into the portfolio of experimentalists working with quantum materials as well as with quantum simulators to benchmark their properties.

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