Experimental determination of thermal entanglement in spin clusters using magnetic susceptibility measurements

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The present work reports an experimental observation of thermal entanglement in a clusterized spin chain formed in the compound $Na_2Cu_5Si_4O_{14}$. The presence of entanglement was investigated through two measured quantities, an entanglement witness and the entanglement of formation, both derived from the magnetic susceptibility. It was found that pairwise entanglement exists below $\sim\!200$ K. Tripartite entanglement was also observed below $\sim\!240$ K. A theoretical study of entanglement evolution as a function of applied field and temperature is also presented.

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I. INTRODUCTION

Since the early years of quantum mechanics, entanglement has attracted much attention due to its fascinating features, such as nonlocality, as exemplified in the Einstein, Podolsky, and Rosen paradox. Recently, it has been discovered that entangled states constitute a valuable resource for quantum information processing and it has raised a great number of studies about entanglement in many different quantum systems. Over the past few years, much effort has been done in developing methods to detect and quantify entanglement.

Until a few years ago, entanglement was not believed to exist beyond atomic scale. The most common arguments against entanglement on larger scales is that macroscopic objects contain a large number of constituents that interact with its surroundings, inducing the decoherence phenomena which leads to the loss of entanglement as size and complexity, and the system's temperature increases. Surprisingly, it was theoretically demonstrated^{3,4} that entangled states can exist in solids at finite temperature and this kind of entanglement is referred in literature as "thermal entanglement." Since the publication of several theoretical works,^{3–20} a few experimental evidences have been reported,^{21–24} confirming the presence of entanglement in solids state systems.

The study of entanglement in solid state physics (for a detailed review, see Ref. 25) is of great relevance to the area of quantum information and quantum computation since many proposals of quantum chips are solid state based. Furthermore, the demonstration that entanglement can change the thermodynamical properties of solids, such as magnetic susceptibility,²¹ shows that entanglement can be related to significant macroscopic effects. Hence, this subject establishes an interesting connection between quantum information theory and condensed matter physics.

The task of entanglement quantification is still an open problem in a general case (for recent reviews, see Refs. 26–28). Many quantities have been proposed to quantify entanglement, one of them is the Entanglement of Formation (EF),^{29,30} which can be easily calculated in the case of two spin 1/2 particles, but it cannot be measured directly in most of the cases. Hence, usually, the detection of entanglement is

done using a quantity called entanglement witness (EW),³¹ which is an observable that, by definition, has a positive expectation value for separable states and negative for some entangled states.

In this work, we report an experimental observation of thermal entanglement in the spin chain system formed in the compound $\mathrm{Na_2Cu_5Si_4O_{14}},^{32,33}$ by using a thermodynamical EW based on the magnetic susceptibility. Furthermore, we have also derived, using the complete knowledge of the Hamiltonian of the spin chain, a relationship between the entanglement of formation of each pair of spins and the experimental magnetic susceptibility, which allowed the determination of the EF directly from the experimental data.

This paper is organized as follows. In Sec. II, we give a brief description of the particular system studied here. The following sections, Secs. III and IV, contain the experimental results at zero applied field and a theoretical study of the entanglement evolution as a function of temperature and applied field, respectively. In the last section, some comments and conclusions are drawn.

II. SPIN CHAIN DESCRIPTION

The copper atoms in the compound Na₂Cu₅Si₄O₁₄ have S=1/2 and are separated into two groups, containing two and three atoms each, named dimer and trimer, respectively. The whole structure of this compound is comprised of zigzag clusters of copper and oxygen atoms, then forming dimertrimer sets of spins, as shown in Fig. 1. There is an indirect exchange interaction between the spins, through the electronic oxygen clouds, and is of short range order, i.e., mainly between the first neighbors. As previously shown,³² the three spins that form the trimer are coupled antiferromagnetically with each other, whereas the two atoms of the dimer are coupled ferromagnetically. In addition, the two sets of spins, dimer and trimer, interact antiferromagnetically with each other. Therefore, labeling the spins according to Fig. 2, the magnetism of the this system can be described by the Hamiltonian

$$\mathcal{H} = -J_1(\vec{S}_1 \cdot \vec{S}_2 + \vec{S}_2 \cdot \vec{S}_3) - J_2(\vec{S}_A \cdot \vec{S}_B) -J_3(\vec{S}_4 \cdot \vec{S}_5) - g\mu_B \vec{H} \cdot \vec{S},$$
 (1)

where g is the Landé g factor, μ_B is the Bohr magneton, \vec{H} is

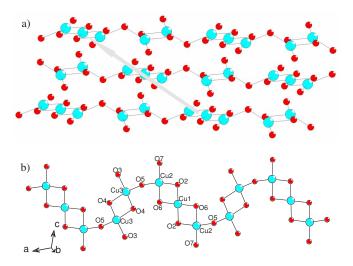


FIG. 1. (Color online) The structure of the $Na_2Cu_5Si_4O_{14}$ compound, the smaller (red) circles being the oxygen atoms and the larger (pale blue) ones the copper atoms, from two different views. (a) Side view, with an arrow indicating the staggered stacking of the chains. (b) Top view of the chain.

an external magnetic field, $\vec{S_A} = \vec{S_4} + \vec{S_5}$ is the total spin of the dimer, $\vec{S_B} = \vec{S_1} + \vec{S_2} + \vec{S_3}$ is the total spin of the trimer, and $\vec{S} = \vec{S_A} + \vec{S_B}$ stands for the total spin of the cluster. The values of the exchange integrals were determined experimentally,³² and they were found to be $J_1 = -224.9$ K, $J_3 = 40.22$ K, and $J_2 = -8.01$ K, for intratrimer, intradimer, and dimer-trimer, respectively.

At finite temperatures T, the thermal equilibrium quantum state of the system is described by the density matrix $\rho(T) = \exp(-\mathcal{H}/k_BT)/Z$, where Z is the partition function. From $\rho(T)$, it is possible to calculate thermodynamical quantities, such as the magnetic susceptibility. Since the Hamiltonian \mathcal{H} commutes with the spin component along the z direction S^z , one can show that the magnetic susceptibility along a given direction α can be written as³⁴

$$\chi^{\alpha}(T) = \frac{(g\,\mu_B)^2}{k_B T} \left(\sum_{i,j=1}^N \left\langle S_i^{\alpha} S_j^{\alpha} \right\rangle - \left\langle \sum_{i=1}^N S_i^{\alpha} \right\rangle^2 \right). \tag{2}$$

In Fig. 3, the theoretical prediction of Eq. (2) is compared to the experimental magnetic susceptibility measured using a conventional superconducting quantum interference device magnetometer with an applied field of 100 Oe as a function of temperature. As can be seen from the figure, there is a monotonic increase of susceptibility down to 8 K, where there is a sharp drop associated with a transition to a three-

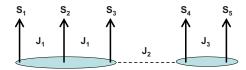


FIG. 2. (Color online) Schematic representation of the dimertrimer cluster and the respective interactions (J_i) between the Cu atoms, according to Eq. (1).

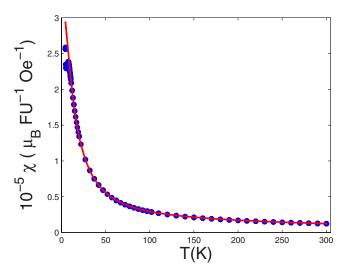


FIG. 3. (Color online) Magnetic susceptibility as a function of temperature with an applied field of 100 Oe. The (blue) points (●) are the experimental results and the (red) solid line is the theoretical prediction based on Eq. (2).

dimensional (3D) ground state.³² The good agreement between the calculations and the experimental data above 8 K shows the faithful of the dimer-trimer cluster model described by Hamiltonian (1).

III. ENTANGLEMENT FOR H=0

In this section, two quantities are used to investigate the presence of entanglement: an entanglement witness and the entanglement of formation, both obtained from the magnetic susceptibility. This study was carried out for the whole system and also for subsystems comprised of two and three spins.

A. Magnetic susceptibility witness

The concept of entanglement witness was first introduced by Horodecki *et al.*,³¹ where it was defined that an EW is an observable which is capable of identifying if a system is in an entangled state. However, in most of cases, an EW cannot quantify the amount of entanglement present in the system and it gives only a sufficient condition for the presence of entanglement. In other words, an EW can only tell if, for some condition, a given system is in an entangled state, but if this condition is violated, we cannot state with absolute certainty that the system's state is separable.

Up to now, various thermodynamical quantities, such as magnetization, ¹⁹ internal energy, ^{13–17,19,20} and heat capacity, ²⁰ have been proposed as an EW, but these depend on the complete knowledge of the specific model describing the system and usually are not directly measurable quantities. However, recently, it has been demonstrated, by Wieśniak *et al.*, ³⁴ that the magnetic susceptibility can be used as an EW for a large class of systems. This witness can be measured directly and can be applicable, in principle, without the full knowledge of the model Hamiltonian. If a system, which has the symmetry $[\mathcal{H}, S^z] = 0$, is in an entangled state, then

the average of the magnetic susceptibility $\bar{\chi}^{expt}(T)$ measured along the three orthogonal axis satisfies the relation³⁴

$$\bar{\chi}^{expt}(T) = \frac{\chi^{x}(T) + \chi^{y}(T) + \chi^{z}(T)}{3} < \frac{(g\mu_{B})^{2}NS}{3k_{B}T},$$
 (3)

where N is the number of spin-S particles. From Eq. (3), we can define the entanglement witness as

$$EW(N) = \frac{3k_B T \overline{\chi}^{expt}(T)}{(g\mu_B)^2 NS} - 1.$$
 (4)

Thus, systems with EW(N) < 0 are in an entangled state. However, it must be emphasized that the condition EW(N) ≥ 0 does not necessarily implies separability. With EW(5) obtained from the measured magnetic susceptibility, we can determine the presence of entanglement in our five spins system. However, it is also interesting to study if the subsystems are in an entangled state. In order to analyze the entanglement between spins in a subsystem, it is necessary to derive the contribution of the susceptibility $\bar{\chi}_{sub}(T)$ due to only the spins that matter. Theoretically, this can be done according to Eq. (2), but using the reduced density matrix $\rho_{sub}(T)$, which represents the density matrix of the subsystem. The $\rho_{sub}(T)$ can be obtained by using the partial trace operation,² which sums over all the possible states of the spins in the system, except those belonging to the subsystem. From the numerical calculation of the total susceptibility and subsystem's contribution, we can define the ratio $R_{sub}^{theor}(T) = \overline{\chi}_{sub}^{theor}(T) / \overline{\chi}^{theor}(T)$ which represents the fraction of the subsystem's contribution to the total magnetic susceptibility. With this quantity at hand, which can only be calculated with the knowledge of the Hamiltonian, it is possible to extract separately, from the experimental data, which contains the contribution of all spins, the magnetic susceptibility of the trimer $\bar{\chi}_{tri}^{expt}(T) = R_{tri}^{theor}(T) \times \bar{\chi}^{expt}(T)$, dimer $\bar{\chi}_{dim}^{expt}(T) = R_{dim}^{theor}(T) \times \bar{\chi}^{exp}(T)$, and

In Fig. 4, we show the experimental data; as can be seen, the EW(5) extracted from the total magnetic susceptibility is negative for any temperature below ~ 110 K, showing the presence of entanglement in the system. Furthermore, the fact that EW(3) for the trimer is negative below ~ 240 K and EW(2) for the dimer is always positive suggests that entanglement only occurs between spins inside the trimer.

B. Entanglement of formation

The entanglement of formation, 29,30 which can be seen as the amount of quantum resources needed to create a given entangled state, is one of the most used for entanglement measurement. This was proposed to quantify the entanglement of a bipartite system. Unfortunately, as many others proposals, the EF is extremely difficult to calculate, in general. However, in the special case of two spin 1/2 particles, recently, an analytical expression was derived by Wootters who showed that the EF of any density matrix (ρ) of two spin 1/2 particles is given by 30

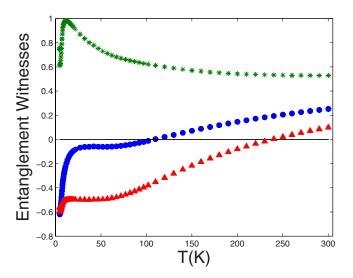


FIG. 4. (Color online) Entanglement witness for the total system EW(5) (blue) circles (\bullet), for the trimer EW(3) (red) triangles (\blacktriangle), and for the dimer EW(2) (green) stars (\star).

$$EF = -x \log_2(x) - (1 - x)\log_2(1 - x), \tag{5}$$

where $x=(1+\sqrt{1-C^2})/2$, C being the concurrence defined as $\max(0,\sqrt{\Lambda_1}-\sqrt{\Lambda_2}-\sqrt{\Lambda_3}-\sqrt{\Lambda_4})$, and Λ 's are the eigenvalues of $R=\rho\sigma_y\otimes\sigma_y\rho^*\sigma_y\otimes\sigma_y$, labeled in decreasing order. The degree of entanglement varies from 0 to 1. A pair of spins is considered to be in a maximally entangled state if EF=1 and separable when EF is equal to zero. For any other values, the state of the spins is said to be partially entangled.

Since Hamiltonian (1) commutes with S^z , we can write the reduced density matrix of the spins located in sites i and j as 13,35

$$\rho_{ij}(T) = \begin{pmatrix} u^+ & 0 & 0 & 0 \\ 0 & \omega_1 & z^* & 0 \\ 0 & z & \omega_1 & 0 \\ 0 & 0 & 0 & u^- \end{pmatrix}, \tag{6}$$

where $u^{\pm} = (1 \pm 2\langle S_i^z + S_j^z \rangle + 4\langle S_i^z S_j^z \rangle)/4$ and $z = \langle S_i^x S_j^x \rangle + \langle S_i^y S_j^y \rangle + i\langle S_i^x S_j^y \rangle - i\langle S_i^y S_j^x \rangle$. The concurrence of such density matrix can be written as

$$C = 2 \max(0, |z| - \sqrt{u^+ u^-}). \tag{7}$$

Now, exploring the fact that the system is isotropic at zero magnetic field, it is possible to write $\langle S_i^x S_j^x \rangle = \langle S_i^y S_j^y \rangle = \langle S_i^z S_j^z \rangle =$

$$C_{ij}(T) = \frac{k_B T}{(g\mu_B)^2} \max\left(0, 2\left|\overline{\chi}_{ij}^{expt}(T) - \frac{(g\mu_B)^2}{2k_B T}\right| - \overline{\chi}_{ij}^{expt}(T)\right). \tag{8}$$

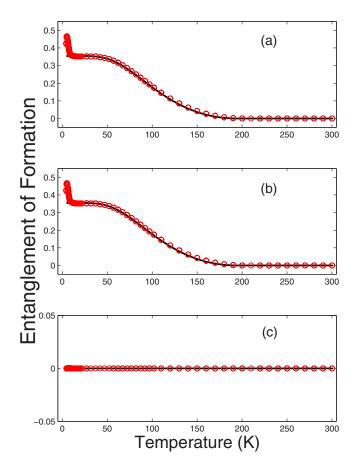
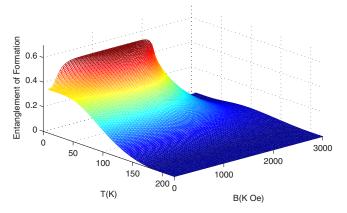


FIG. 5. (Color online) The experimentally determined entanglement of formation, the (red) circles, for the pairs inside the trimer (a) 1-2, (b) 2-3, and (c) 1-3. The (black) solid lines are the theoretical prediction. The small discrepancy between theory and experiment at low temperatures is associated with a 3D transition (Ref. 32).

Equation (8) relates the concurrence of the pair i-j, hence the EF, to the magnetic susceptibility $\overline{\chi}_{ij}^{expt}(T)$, which can be obtained from the experimental data, as explained before. It is interesting to note that Eq. (8) leads to the concurrence found by Asoudeh and Karimipour⁹ for mean-field clusters.

In Fig. 5, we show the experimental EF obtained from the measured magnetic susceptibility and the theoretical EF calculated with the reduced density matrix $\rho_{ij}(T)$ for the pairs inside the trimer (1-2, 2-3, and 1-3). We can see that there is entanglement only between the pairs of spins 1-2 and 2-3 which persists up to a critical temperature $T^c \sim 200$ K. The entanglement of formation for other pairs is always null and is not shown. These results confirm that only the spins in the trimer are entangled, those in the dimer being in a separable state. An interesting feature is that the entanglement witness for the trimer EW(3) gives $T^c \sim 240$ K, while the T^c obtained from the entanglement of formation is ~ 200 K. Since the EF can only see bipartite entanglement, this result indicates that pairwise entanglement is not the sole entanglement in the system and a tripartite entanglement is also present in the trimer.

It is also interesting to ask if this multipartite entanglement is genuine or not. Genuine multipartite entanglement



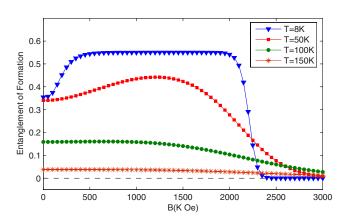
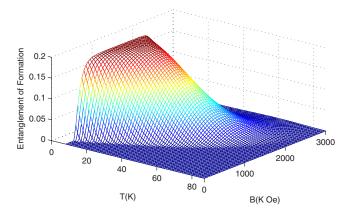


FIG. 6. (Color online) (Top) Entanglement of formation (EF) for pairs 1-2 and 2-3 as a function of temperature and applied field in the *H-T* plane. (Bottom) EF for selected temperatures as a function of applied magnetic field.

can be identified by the criterion described in Ref. 18. Applied to the trimer system, the criterion states that if the expression $\langle \mathcal{H}_{tri} \rangle < J_1(1+\sqrt{5})/4$ holds, where $\langle \mathcal{H}_{tri} \rangle$ is the mean value of the energy corresponding just to the trimer part, then the tripartite entanglement is indeed genuine. We have numerically calculated, from the reduced density matrix of the trimer, the threshold temperature below which the genuine tripartite entanglement exists. We found that the threshold temperature is ~ 108 K. This result suggests that the compound has a genuine tripartite entanglement for a finite temperature. Therefore, this genuine entanglement does not extend up to 240 K. It is also important to emphasize the fact that entanglement is confined in the trimer and thus it is not a macroscopic entanglement but rather a thermal entanglement.

IV. EFFECT OF THE MAGNETIC FIELD

In order to investigate the effect of the application of a magnetic field, the EF was calculated for each pair of spins. In Figs. 6 and 7 we show the EF for the three pairs of spins in the trimer. The entanglement of pair 1-2 behaves the same way as that of pair 2-3; the application of a low magnetic field increases the amount of entanglement of both pairs at



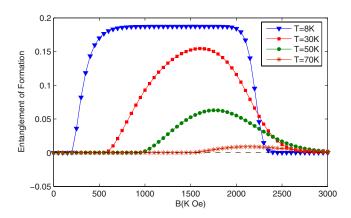


FIG. 7. (Color online) (Top) Entanglement of formation (EF) for pair 1-3 as a function of temperature and applied field in the *H-T* plane. (Bottom) EF for selected temperatures as a function of applied magnetic field.

low temperatures and also creates a small entanglement between pair 1-3, which was zero at zero field. Similar behavior was also encountered in other theoretical works^{4,9} and can be understood in terms of changes in the ground state. At H=0, the ground state of the system is degenerated, being a statistical mixture of two states. This mixture only has entanglement between pairs 1-2 and 2-3. The application of an external magnetic field changes the energy eigenvalues, leading to a different ground state which has a different degree of entanglement.

For a high enough magnetic field, the ground state becomes $|\uparrow\uparrow\uparrow\uparrow\uparrow\rangle$, which corresponds to all spins aligned parallel with the external field. At low temperatures, where the ground state is very populated, EF vanishes suddenly near ~ 2200 kOe due to the sudden change in the ground state to $|\uparrow\uparrow\uparrow\uparrow\uparrow\rangle$. However, for higher temperatures, there are many states populated and the decreasing of entanglement is slow.

From the H-T diagram shown in Figs. 6 and 7, we can also observe that the entanglement of pairs does not occur above the critical temperature $T^c \sim 200$ K and above the critical field $H^c \sim 3000$ kOe. An interesting feature is that the critical field H^c is much higher than the available field intensities that usually can be created in laboratories, which indicates that the entanglement in this system cannot be destroyed by a common magnetic field for temperatures below 200 K, providing that high magnetic fields do not induce any structural changes in the compound. The entanglement of formation for other pairs is always zero at any point in the H-T plane and is not shown.

V. CONCLUSIONS

In summary, we have successfully established the presthermal entanglement in the compound Na₂Cu₅Si₄O₁₄. From the obtained results, we could conclude that entanglement is strong in this system and disappears only at high temperatures and high magnetic fields. At zero magnetic field, we found experimentally that pairwise entanglement exists below ~200 K and tripartite entanglement exists below ~240 K, which shows that multipartite entanglement is stronger than bipartite entanglement in our case. The same feature was also verified experimentally in the compound Na₂V₃O₇.²² The entanglement observed here is confined into the small cluster of spin and thus it is not a macroscopic entanglement. We have also presented a theoretical study of the entanglement evolution as a function of applied field and temperature, showing that magnetic field can increase the degree of entanglement in pairs 1-2, 2-3, and

We emphasize that some results of this paper is based on the validity of the dimer-trimer model; however, this model is shown to be a good model for the present system (see Fig. 3). Furthermore, since the entanglement witness EW(5) does not require any assumption about the model or the explicit values of the exchange-coupling parameters, even if the model is not completely correct, the main conclusion does not change, i.e., the presence of thermal entanglement in the compound $Na_2Cu_5Si_4O_{14}$.

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