Experimental detection of quantum information sharing and its quantification in quantum spin systems

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Abstract. We study the macroscopic entanglement properties of a low-dimensional quantum spin system by investigating its magnetic properties at low temperatures and high magnetic fields. The spin system chosen for this is copper nitrate \( (\text{Cu(NO}_3\text{)}_2 \times 2.5\text{H}_2\text{O}) \), which is a spin chain that exhibits dimerization. The temperature and magnetic field dependence of entanglement from the susceptibility and magnetization data are given, by comparing the experimental results with the theoretical estimates. Extraction of entanglement has been made possible through the macroscopic witness operator, magnetic susceptibility. An explicit comparison of the experimental extraction of entanglement with theoretical estimates is provided. It was found that theory and experiments match over a wide range of temperatures and fields. The spin system studied exhibits quantum phase transition (QPT) at low temperatures when the magnetic field is swept through a critical value. We show explicitly for the first time, using tools used in quantum information processing, that QPT can be captured experimentally using quantum complementary observables, which clearly delineate entangled states from separable ones across the QPT.

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We have also estimated the partial information sharing in this system from our magnetization and susceptibility data. The complementarity relation has been experimentally verified to hold in this system.

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1. Introduction

Quantum computation and information has had tremendous appeal to researchers, both on the theoretical and experimental fronts [1, 2]. The most important element of quantum protocols is the quantum correlation arising from entanglement [3–5]. Entanglement in quantum condensed matter systems has been extensively studied from the perspective of quantum information processing over the last decade. Various measures of entanglement and its quantification are now available [6, 7]. In recent years, there has been significant development on the quantification of entanglement, especially in spin systems, where extraction of entanglement has been made possible through macroscopic witness operators such as magnetic susceptibility [8–13]. Spin chains have been proposed for possible applications in making components such as quantum wires, which can be used to connect various quantum registers or gates in a quantum computer. The study of entanglement characteristics of spin chains is important for their application in the implementation of quantum communication protocols [14, 15]. This has led to the study of different properties of Heisenberg spin chains [16–18] that would be used for computations as well as quantum communications [19]. Spin systems, with their finite-dimensional Hilbert spaces, are ideal for studying the dynamics of entangled states. For the purpose of teleportation and other quantum protocols, maximally entangled channels provide quantum information transfer with very high fidelity; hence it is imperative that one should quantify the entanglement of a particular system or channel [20, 21].

In this work, we have studied the magnetic properties of a spin chain compound, copper nitrate (\(\text{Cu(NO}_3\text{)}_2 \times 2.5\text{H}_2\text{O}\)), which exhibits dimerization and gives us an opportunity to study both bipartite entanglement and quantum phase transition (QPT) owing to a reduction in the relevant degrees of freedom, enabling one to capture the dynamics in a reduced Hilbert space. In this system, the Cu\(^{2+}\) has an unpaired, localized electron. It is known to be an alternating dimer spin chain, where the interaction between alternating spins (say, spin numbers 2 and 3) is much less than that between the other two spins (spin numbers 1 and 2). This enables us to model the system with a four-spin Hamiltonian owing to translational invariance. Here, we study the temperature and magnetic field dependence of magnetic susceptibility and magnetization. We subsequently extract the temperature and magnetic field dependence of entanglement from...
the susceptibility and magnetization data. As mentioned above, magnetic susceptibility has been shown to be an entanglement witness (EW) \[8\] and comparisons may be drawn with existing measures of entanglement for mixed states, namely, concurrence and entanglement of formation (EoF). These are bipartite measures of entanglement. The entanglement between any two spins of a spin chain is obtained by tracing out the extra degrees of freedom and using the resultant reduced density matrix to quantify the entanglement. Even though our system is an alternating dimer, where the entanglement between two spins (spins 1 and 2) coupled through the stronger interaction ‘\(J\)’ is maximal at the cost of entanglement between spins 2 and 3. Susceptibility, being the average two-point spin–spin correlation function, captures the average entanglement between any two spins. Thus, to model the entanglement data, we considered only the entanglement between the spins within a dimer, which are maximally entangled and can be represented by a two-qubit system. Thus, entanglement of a two-qubit system is sufficient to compare with the experimentally extracted entanglement by using susceptibility as an EW. We have compared the temperature and field dependence of the entanglement so obtained from our experimental data with the temperature and field variation given theoretically by Arnesen \textit{et al} \[16\], who used a similar dimer model.

These spin systems also exhibit QPT at low temperatures, when the magnetic field is swept through a critical value. We have captured this using the tools of quantum information processing. There have been efforts to unravel the extra correlations present close to a quantum critical point (QCP) invoking concepts from quantum information theory \[22, 23\], especially the non-locality inherent in entanglement. Wieśniak \textit{et al} \[8\] have proposed two macroscopic quantum complementary observables (‘\(P\)’ and ‘\(Q\)’), based on magnetization and susceptibility, that can be used to detect the QPT in dimerized systems. One of these complementary observables (‘\(P\)’) captures the local properties of spins, whereas the other (‘\(Q\)’) captures the non-local features in a quantum spin system. We have used these complementary observables in our study to detect QPT in our spin system. However, since QPT is a many-body effect, one needs to consider the spin chain and not just the two qubits mentioned above to explain our data. Since the system behaves like an alternating dimer where two dimers are weakly coupled by the weaker interaction ‘\(J\)’, translational invariance allows one to restrict oneself to four qubits and hence to a finite-dimensional Hilbert space (16 dimensions), instead of an infinite-dimensional Hilbert space. Due to this reduction, there is a clear crossover of the ground state and the first excited state as a function of magnetic field at low temperatures. We have also demonstrated partial information sharing from our magnetization and susceptibility data using these complementary observables.

The main results of this paper have been presented in two major sections, one devoted to the study of entanglement in this system (section 3) and the other to the study of QPT (section 4). In the following section, we start with reviewing the entanglement property of spin chains, especially pertaining to its quantification through macroscopic EW in the form of susceptibility \[8\]. Subsequently, in section 3, we proceed to the experimental susceptibility of this dimerized spin system and characterization of its magnetic properties. We quantify the entanglement using the EW operator and demonstrate the effect of temperature and magnetic field on entanglement. In section 4, we present the high-field magnetization data demonstrating the observation of QPT. It is observed in the present case that a QPT separates the entangled and non-entangled Hilbert spaces.
2. Entanglement in spin chains

We briefly review the entanglement properties of different quantum states of a spin chain in this section. The Hamiltonian of a spin chain, with nearest-neighbour interaction, is given by

\[ H = J \sum_i \vec{S}_i \cdot \vec{S}_{i+1} + B \sum_i S_z^i, \]

where the spins are given by (in units of \( \bar{\hbar} = 1 \))

\[ S_x^i = \sigma_x^i, \quad S_y^i = \sigma_y^i, \quad S_z^i = \sigma_z^i; \]

and \( \sigma_x^i, \sigma_y^i, \sigma_z^i \) are the three Pauli spin matrices.

The magnetic field is assumed to be applied along the \( \hat{z} \)-direction. The net magnetization would be defined as the expectation value of \( \sigma_z \), the eigenvectors of which are \( |\uparrow\rangle = \left( \begin{array}{c} 1 \\ 0 \end{array} \right) \) and \( |\downarrow\rangle = \left( \begin{array}{c} 0 \\ 1 \end{array} \right) \).

Since most of the entanglement measures currently available are for bipartite systems, it is worth exploring how one can extract the bipartite entanglement from given susceptibility data for a spin chain. Keeping this in mind, we consider a two-spin system, the Hamiltonian for which can be written as

\[ H = J \vec{S}_1 \cdot \vec{S}_2 + B(S_z^1 + S_z^2) = \frac{J}{4} \left( \sigma_x^1 \cdot \sigma_x^2 + \sigma_y^1 \cdot \sigma_y^2 + \sigma_z^1 \cdot \sigma_z^2 \right) + \frac{B}{2} (\sigma_z^1 + \sigma_z^2). \]

The eigenvalues and the corresponding eigenvectors of this Hamiltonian are:

\[ E_1 = \frac{J}{4} + B; \quad |\uparrow\uparrow\rangle = \left( \begin{array}{c} 1 \\ 0 \\ 0 \end{array} \right), \quad E_2 = \frac{J}{4} - B; \quad |\downarrow\downarrow\rangle = \left( \begin{array}{c} 0 \\ 0 \\ 1 \end{array} \right), \]

\[ E_3 = \frac{J}{4}; \quad |\psi^+\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle) = \frac{1}{\sqrt{2}} \left( \begin{array}{c} 1 \\ 1 \\ 0 \end{array} \right), \]

\[ E_4 = -\frac{3J}{4}; \quad |\psi^-\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) = \frac{1}{\sqrt{2}} \left( \begin{array}{c} 0 \\ 1 \\ -1 \end{array} \right), \]

where \( |\psi^+\rangle \) and \( |\psi^-\rangle \) are the Bell states.

A ferromagnetic system \((J < 0)\) in the absence of an external magnetic field does not display any entanglement in the ground state. This is because \( \hat{S}_i^z \) commutes with the Hamiltonian; the rate of change of the order parameter, the expectation value of \( \hat{S}_i^z \), is zero. Therefore, there is no spin fluctuation at low temperatures. In comparison, for an antiferromagnetic ground state \((J > 0)\), the order parameter, which is the staggered magnetization, is defined as \( \langle M_s \rangle = \frac{1}{N} \left( \sum_i (-1)^i S_i^z \right) \). As \( \frac{1}{2} \left( (-1)^i S_i^z + (-1)^{i+1} S_{i+1}^z \right) \) does not commute with the Hamiltonian, there will be spin fluctuation even at zero temperature \( \frac{dM_s}{dt} = \langle \hat{H}, \hat{S}_i \rangle \neq 0 \). Hence, this system is entangled in the absence of an external field. In fact, the ground state of an antiferromagnetic interaction \((J > 0)\) is the maximally entangled state \((|\psi^-\rangle)\) for the above system, whereas the ground state for \( J < 0 \) is an equal mixture of the three-fold degenerate triplet states and is a separable state.
For the above case, the staggered magnetization is the order parameter and the magnetic susceptibility, which is the fluctuation of this order parameter, can be used as an EW. An EW is an observable whose expectation value is positive for an entangled state, although a negative expectation value does not ensure separability [7]. Magnetic susceptibility has been shown to be a macroscopic EW [8], where the authors show that susceptibility is a measure of entanglement, a non-local property of the spins. Its canonically conjugate observable, magnetization is a measure of the local spin properties and is therefore an observable complementary to the magnetic susceptibility. As the magnetic susceptibility can be measured experimentally, it is possible to extract a measure of entanglement from the experimental data [24, 25]. One of the first systems that exhibited entanglement through bulk measurements such as magnetic susceptibility or heat capacity is the insulating magnetic salt LiHo$_x$Y$_{1-x}$F$_4$ [26].

To extract the entanglement and to compare with the theoretical values, one needs to consider the measure given by Wootters [27], where it has been shown that concurrence $C$ is a good measure of entanglement,

$$C = \max(0, \sqrt{\lambda_1} - \sqrt{\lambda_2} - \sqrt{\lambda_3} - \sqrt{\lambda_4}),$$  \hspace{1cm} (3)

where $\lambda_1 \geq \lambda_2 \geq \lambda_3 \geq \lambda_4$ are the eigenvalues of the operator,

$$\tilde{\rho}_{12} = \sigma_2 \otimes \sigma_2 \rho_{12}^\ast \sigma_2 \otimes \sigma_2.$$  \hspace{1cm} (4)

$\rho_{12}$ is the two-particle reduced density matrix and the asterisk denotes complex conjugation. It has been shown by O’Connor and Wootters [28] that, for an antiferromagnet, the concurrence is given by

$$C = \frac{1}{2} \max[0, \frac{|U|}{NJ} - 1].$$  \hspace{1cm} (5)

in the absence of a magnetic field. Here, $U$ is the free energy, $U = \langle H \rangle$. Thus, considering the system to be isotropic, the formula for concurrence, given in equation (5), can be written as

$$C = 2 \max[0, \langle \vec{S}_1 \cdot \vec{S}_2 \rangle - (1/4)].$$  \hspace{1cm} (6)

This $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$ will be related to magnetic susceptibility in the next section, where we extract the entanglement out of experimental susceptibility data.

3. Experimental results

The system that we have considered for this paper is copper nitrate (Cu(NO$_3$)$_2$ $\times$ 2.5H$_2$O), an inorganic compound and a spin-$\frac{1}{2}$ system procured from Sigma-Aldrich. Cu(NO$_3$)$_2$ $\times$ 2.5H$_2$O has been shown to be an alternative dimer spin chain [29]. The corresponding spin Hamiltonian is of the type

$$H = \sum_i (J_1 S_{2i} \cdot S_{2i+1} + J_2 S_{2i+1} \cdot S_{2i+2}),$$  \hspace{1cm} (7)

representing pairs of spins that are alternately coupled by strong intra-dimer coupling $J_1$ and weak inter-dimer coupling $J_2 \approx 0.25 \ J_1$ [29]. This system behaves like a chain of dimers or exchange coupled pairs, which are themselves weakly coupled with each other.

The experiment was performed on single crystals of copper nitrate having a purity of 99.999%. Being very sensitive to exposure to air, the crystals were preserved inside a refrigerator and were transported in liquid nitrogen before measurement. Static magnetic susceptibility and
isothermal magnetization measurements were made in a Quantum Design magnetic property measurement system (MPMS). The static magnetic field is applied using a superconducting magnet. Susceptibility data were taken in the temperature range 2–20 K. Magnetization isotherms were taken as a function of field at various temperatures. The field was varied from 0 to 7 T and the temperature was varied from 2 to 9 K. The ac susceptibility measurement was also made in the temperature range 2–9 K with a very small oscillating excitation field with an amplitude of 3 Oe. The frequency of the oscillating field was varied from 0.1 to 1500 Hz. There was no perceptible frequency dependence as is expected for an antiferromagnet, indicating the absence of any disorder or ferromagnetic phase. A proper minimization of trapped magnetic field was carried out before commencement of every measurement. Since magnetic susceptibility is defined as the field derivative of magnetization, we verified that the magnetic susceptibility obtained by taking the field derivatives of experimental magnetization isotherms was in agreement with the ac susceptibility data measured at different fields and temperatures. The temperature dependence of susceptibility in zero field is shown in figure 1(a).

The magnetic susceptibility, \( \chi \), can be defined as the field derivative of magnetization \( M \) (in the applied field direction, which here is \( \hat{z} \)) in the limit \( B \to 0 \), and the magnetization, \( M \), is defined as the sum of the expectation values of the \( \hat{z} \)-component of individual spins, \( M = \sum_{i=1}^{N} \langle \sigma_i^z \rangle \). The expression for susceptibility derived from an alternative dimer model is given as [24, 25]

\[
\chi = \left( \frac{\delta M}{\delta B} \right)_{B=0} = (g^2 \mu_B^2 / k_B T) \langle M^2 \rangle_{B=0} = (g^2 \mu_B^2 / k_B T) \sum_i \langle S_i^z S_{i+1}^z \rangle \\
\approx (g^2 \mu_B^2 N / k_B T) \left[ (1/4) + \langle \vec{S}_1 \cdot \vec{S}_2 \rangle / 3 \right],
\]

(susceptibility data (circles) of Cu(NO$_3$)$_2 \times 2.5$H$_2$O and fit (solid line) to equation (8), corresponding to the alternating dimer model (equation (8)). The solid green line delineates the separable regime (equation (12)) from the entangled regime (see text). (b) Extraction of entanglement from the susceptibility data of copper nitrate using equation (10). The solid line is the theoretical value of entanglement calculated using equation (11), given for comparison to the experimentally extracted data.

**Figure 1.** (a) Temperature (K) vs. susceptibility (emu/mol) for Cu(NO$_3$)$_2 \times 2.5$H$_2$O. (b) Temperature (K) vs. entanglement.
where $N$ is the total number of spins per mole, $\mu_B$ is the Bohr magneton and $k_B$ is the Boltzmann constant. Here, it was assumed that the exchange term and the Zeeman term of the Hamiltonian commute with each other, i.e. $[J \sum \vec{S}_i \cdot \vec{S}_j, B \sum_i S_i^z] = 0$. The expression in equation (8) was derived using the isotropy of space, nearest-neighbour interaction and the fact that $(\vec{S}_1 + \vec{S}_2) = 0$ for an antiferromagnet. One can evaluate $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$, from the bipartite Hamiltonian (equation (2)), for the $B = 0$ limit, yielding

$$\langle \vec{S}_1 \cdot \vec{S}_2 \rangle = \left( -\frac{3}{4} \right) \frac{1 - e^{-\frac{\pi}{\tau}}}{1 + 3 e^{-\frac{\pi}{\tau}}}.$$  

(9)

The zero-field susceptibility data were fitted to the formula for susceptibility given in equation (8) having substituted the above expression for $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$. The exchange constant $J$ is treated as a fitting parameter and yielded a value of $'J/k_B = 4 \text{ K}'$. The fitting is depicted in figure 1(a). The solid line shows the fitted curve. The data (open circles) and the fit (solid line) are in good agreement with each other.

Now, combining equation (6) and the expression $\chi = (g^2 \mu_B^2 / k_B T) \sum_i (S_i^z S_{i+1}^z)$, given in equation (8), one obtains the expression for EW in terms of susceptibility,

$$C = \max \left[ 0, 1 - \frac{(6k_B T \chi)}{g^2 \mu_B^2 N} \right].$$  

(10)

which is consistent with that introduced by Bruss [30]. This expression for EW is similar to the theoretical measure of concurrence. Upon further substituting $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle = \left( -\frac{3}{4} \right) \frac{1 - e^{-\frac{\pi}{\tau}}}{1 + 3 e^{-\frac{\pi}{\tau}}}$ from equation (9), one obtains the expression for EW,

$$C = 2 \max \left[ 0, \frac{1 - e^{-\frac{\pi}{\tau}}}{1 + 3 e^{-\frac{\pi}{\tau}}} \right].$$  

(11)

For a separable state, the magnetic susceptibility will satisfy

$$\chi \geq \frac{g^2 \mu_B^2 N}{k_B T} \frac{1}{6}.$$  

(12)

Entanglement in Cu(NO$_3$)$_2 \times 2.5\text{H}_2\text{O}$ has been reported earlier in a theoretical paper by Brukner et al [24], who used the susceptibility data of Berger et al [31] to extract the entanglement. It is worth pointing out that EW, which in spin systems is susceptibility, provides a more general quantification of entanglement and hence could be applicable to a general spin chain, with spin-$\frac{1}{2}$, spin-1 or spin-$\frac{3}{2}$. However, concurrence is defined only for the spin-$\frac{1}{2}$ system [27]. We make use of the expression for EW given in equation (10) to quantify the entanglement content from the experimental data as outlined below.

The dotted line shows the entanglement boundary given by equation (12); the entangled region is represented by the region to the left of the dotted line. The copper spins exhibit entanglement up to the temperature where antiferromagnetic correlations persist, i.e. up to around 5 K. This is slightly more than the ordering temperature, which corresponds to the peak in the susceptibility curve. Figure 1(b) shows the extracted value of entanglement from the experimental data (open circles), which were extracted using the expression for EW (concurrence) given in equation (10). The comparison of the theoretically estimated values (solid line) to the experimentally extracted data is also shown here. It was generated using the expression for concurrence given in equation (11) for $J = 4 \text{ K}$. It can be seen that the entanglement vanishes above the temperature where the antiferromagnetic correlations would
vanish. Here too, the calculated theoretical values (solid line) match reasonably well with the experimentally extracted values of entanglement, generated using the EW (figure 1(b)). Thus, entanglement is not only an entity which is quantifiable, but also can be measured experimentally.

The effect of temperature and magnetic field on the entanglement of a Heisenberg antiferromagnet, considering the dimer model, has been dealt with theoretically by Arnesen et al [16], wherein they consider the effect of temperature-induced magnons on entanglement. In figure 2(a) we re-generated the theoretical three-dimensional (3D) plot given in figure 1 of Arnesen et al [16]; however, unlike theirs, which was plotted in arbitrary units, here both the temperature and the magnetic field have been scaled exactly to match with experimental data. The magnetic field values are in units of Tesla and the temperature is in kelvin. The entanglement (concurrence) in this plot was generated for the dimer model, considering $J = 4\text{ K}$. The experimental 3D plot of concurrence as a function of field and temperature is shown in figure 2(b).

There is a remarkable similarity between the theoretical and experimental 3D plots. Although we have collected magnetization data up to 7 T, we have shown the experimental plot only up to 4 T. This is partly because most of the relevant physics pertaining to this system is captured up to 4 T, and also because of some subtleties involved in the analysis of susceptibility data above 4 T, which is mentioned in the next section. One can see that the general pattern of decreasing entanglement with increasing temperature is consistent in both the theoretical and experimental plots, as is the decrease in entanglement with an increase in magnetic field at the lowest measured temperature. This is due to the greater contribution from the separable triplet state in the statistical mixture as the temperature is increased at a particular field or as the magnetic field is increased at a particular temperature (see figure 2). The plot also shows that the entanglement vanishes at the same temperature (around $T = 5\text{ K}$) irrespective of the strength of the magnetic field.
In figure 3, we have shown the plot of the experimental value of entanglement as a function of magnetic field at $T = 2$ K. Also shown in this figure is the theoretical entanglement as a function of field. As expected, this shows a gradual decrease from the maximum value. To understand this let us consider the following. At a finite temperature, say $T = 2$ K, the state is in a mixture of the four eigenstates; the thermal density matrix of this state is given as

$$\rho = \frac{1}{Z} |\phi^-\rangle \langle \phi^-| e^{\frac{3J}{4} \beta} + |\phi^+\rangle \langle \phi^+| e^{-\frac{3J}{4} \beta} + |\uparrow\uparrow\rangle \langle \uparrow\uparrow| e^{-\frac{(J-B)\beta}{4}} + |\downarrow\downarrow\rangle \langle \downarrow\downarrow| e^{\frac{(J+B)\beta}{4}},$$

(13)

where $\beta = \frac{1}{k_B T}$ and $Z = \text{Tr}(\rho) = e^{\frac{3J}{4} \beta} + e^{-\frac{3J}{4} \beta} + e^{\frac{(J+B)\beta}{4}} + e^{\frac{(J-B)\beta}{4}}$.

At a finite non-zero temperature the system is in a statistical mixture of all the eigenstates. However, at temperatures as low as 2 K, two lowest energy states $|\psi^-\rangle$ and $|\downarrow\downarrow\rangle$ will have the dominant contribution. With an increase in magnetic field, the contribution from a separable state $|\downarrow\downarrow\rangle$ increases gradually at the expense of the contribution from $|\psi^-\rangle$, which is a maximally entangled state. As one keeps on increasing the magnetic field, the $|\downarrow\downarrow\rangle$ state ought to cross the $|\psi^-\rangle$ state and become the lowest energy state above a critical field. Till the QCP, the eigenstates do not change, although the eigenvalues would. At zero temperature the ground state would remain intact below QCP, but for finite temperature we would have a statistical mixture, skewed in favour of the two states mentioned above, closing the excitation gap between the two states. However, despite the closing of this gap, one can still see signatures of QPT at finite temperatures, the detailed analysis of which is given in the next section.

In figure 3, we can see that the experimental variation of concurrence at a fixed temperature is in reasonably good agreement with the theoretical curves, especially at low magnetic fields. The difference between the theoretical and experimental curves at high fields (above 3 T) is probably due to a difference in $J$ in the two scenarios. This discrepancy arises because the theoretical model considered by Arnesen et al [16] is a dimer model (with $J = 4$ K).
whereas Cu(NO$_3$)$_2 \times 2$H$_2$O is an alternative dimer system corresponding to equation (7), with $J_2 = 0.25 J_1$. In addition to that, the isotropy of the system, i.e. the fact that $\chi_x = \chi_y = \chi_z$, breaks down in the case of high magnetic field. However, since this is a dimerized system, leading to the formation of singlet states, where the net $\vec{S} = 0$, there is perfect SU(2) symmetry, as the system is described by an isotropic Heisenberg model, one can use the formula for EW up to a field of 4 T for a system where $J \approx 4$ K.

4. Quantum phase transition

The QPT in a many-body quantum spin system takes place when one sweeps an external physical parameter, such as magnetic field or pressure, at a fixed temperature (near zero temperature) and the nature of the ground state undergoes a qualitative change, such that the symmetry of the ground state changes. In certain types of QPT, as one sweeps the field, a level crossing occurs between the ground state and the excited state, such that an excited state becomes the new ground state.

In the Heisenberg Hamiltonian, given in equation (1), $H(B)$, the exchange term ($H_0 = J \sum \vec{S}_i \cdot \vec{S}_j$) and the Zeeman term ($H_1 = B \sum S_z^i$) commute with each other ($[H_0, H_1] = 0$). Thus $H_0$ and $H_1$ can be simultaneously diagonalized and so the eigenfunctions are independent of the field $B$ even though the eigenvalues vary with $B$. If one allows for the evolution of the ground state energy of $H(B)$ as a function of $B$, with the field being swept adiabatically, then there can be a level crossing where an excited level becomes the ground state at $B = B_c$ (say), creating a point of non-analyticity of the ground state energy as a function of $B$. In the thermodynamic limit or the infinite lattice limit, this is termed a quantum phase transition (QPT). The phase transition is usually accompanied by a qualitative change in the nature of the ground state as one varies the magnetic field, say a crossover from a singlet-like state to a triplet-like ground state [32]. This can happen either in a transverse field Ising model [32] or in an isotropic Heisenberg model where dimerization occurs [33]. The point where the crossover takes place is called the quantum critical point (QCP). The behaviour of entanglement at the QCP has been extensively studied theoretically by several groups [22, 23, 34]. They found that the entanglement scales close to the QCP. This means that the entanglement, which in turn is related to the correlation function, is scale invariant close to the QCP. The QPT in dimer-like systems is characterized by the crossing of energy eigenstates as a function of some parameter such as the magnetic field. This scenario is depicted in figure 4(a) for a dimer system (two-qubit system), where the state $|\uparrow\uparrow\rangle$ crosses the ground state at some critical field and replaces it as the ground state, thereby changing the symmetry of the ground state. Thus the ground state changes from a maximally entangled state to a separable state for a two-qubit system. The exchange interaction for this system $J/k_B = 4$ K. This is chosen to suit copper nitrate.

It is often not possible to go down in the vicinity of $T = 0$ and one has to measure or describe thermodynamic properties at finite temperature. One can understand this by performing expansions about the $T = 0$ QCP and obtain information which is otherwise inaccessible [32]. Magnetization and susceptibility are bulk measurements even though the spins in the chain dimerize; there are $10^{23}$ such dimers, which represents the thermodynamic limit. However, to capture the many-body effect, we did our analysis using the alternating dimer model where two dimers are weakly coupled by the weaker interaction $J_2$ (cf equation (7)). Owing to translational invariance, the system can be aptly described by four qubits/spins, allowing us to restrict ourselves to a finite-dimensional Hilbert space, instead of an infinite-dimensional space.

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The Hamiltonian for such a four-qubit system is given by

\[ H = J_1 \vec{S}_1 \cdot \vec{S}_2 + J_2 \vec{S}_2 \cdot \vec{S}_3 + J_3 \vec{S}_3 \cdot \vec{S}_4 + B(S^z_1 + S^z_2 + S^z_3 + S^z_4), \]

where \( J_1 \) and \( J_2 \) are the two exchange interactions, with \( J_1 \) being significantly larger than \( J_2 \). The energy eigenvalues for this Hamiltonian have been plotted as a function of field for \( J_1/k_B = 4 \text{ K} \) and \( J_2 = 0.25J_1 \). This is the reported value of \( J_1/J_2 \) [29]. One can see that there are two crossovers in the ground state, as depicted in figure 4(b). Thus the system will have a lower critical field \( (B_{C1}) \) of around 3.7 T and an upper critical field \( (B_{C2}) \) of 4.4 T. However, to distinguish between the two critical points one needs to go down to extremely low temperatures, say 50 mK or less. Hence, the system can be represented by an effective J which will capture the essential physics. An easy way to visualize the passing of the dimer model to an alternating dimer model is that in the dimer model, when the field is applied, the three-fold degenerate triplet state splits due to the Zeeman term (figure 4(a)), but the exchange between the dimers broadens the triplet bands giving rise to gradual long-range magnetic order above the critical field, where the triplet band crosses the singlet band [35]. At \( B_{C1} \), the triplon gap closes and the system is driven into an ordered state, with uniform magnetization parallel to the field and antiferromagnetic order perpendicular to the field, or a canted X–Y antiferromagnetic state. This is also known as Bose–Einstein condensation of triplons [36]. Then at \( B_{C2} \), the system enters the fully polarized state.

In a significant effort to understand the quantum correlations close to a QPT, concepts from quantum information processing, especially that of entanglement, were invoked [23]. This was done especially to extract the extra correlations present in QPT, which do not have a classical counterpart. It was found that, close to the critical point, the entanglement depends strongly on the magnetic field. Now since entanglement measures the non-local effect in the system, we also tried to capture the non-locality in the spin system close to QCP.

Here we present a detailed analysis of the magnetic field dependence of magnetization and susceptibility. It has been reported by Wieśniak et al [8] that magnetization and susceptibility are complementary variables. Thus, whereas magnetization captures the local properties of

Figure 4. (a) Energy eigenvalues versus magnetic field for a dimer system. (b) Energy eigenvalues versus magnetic field for an alternating dimer system.
spins, susceptibility exhibits non-local features in a quantum spin system (as is evident from its relation with correlation function). The non-local nature of entanglement intuitively points to the connection between entanglement and susceptibility (cf equation (10)), which in turn is connected with spin–spin correlation functions.

Wieśniak et al [8] have derived a macroscopic quantum complementarity relation, which, in their notation, is given by

$$1 - \frac{kT \tilde{\chi}}{NS} + \frac{(\tilde{M})^2}{N^2S^2} \leq 1. \quad (15)$$

Following their notation, let $Q \equiv 1 - \frac{kT \tilde{\chi}}{NS}$ and $P \equiv \frac{(\tilde{M})^2}{N^2S^2}$, where $P$ describes the local properties of individual spins and $Q$ the non-local quantum correlations between spins. Since $Q$, through its dependence on the susceptibility, is proportional to the two-point correlation function, its positive value implies the existence of entanglement (cf equation (10)). When $Q = 1$, the system is maximally entangled, and in such a case, the local property of spins, $P = 0$. In the other extreme, when $P = 1$ or the magnetization is maximum, the local properties of the spins are well defined at the expense of the non-local property, i.e. $Q = 0$. Thus, mathematically, the above relation describes partial quantum information sharing between local and non-local properties of spins. Following the work of Wieśniak et al, a similar complementary relation was also demonstrated theoretically by Tribedi and Bose [37] for a spin-1 system.

At the heart of this inequality lies the fact that the spin operators, $\tilde{S}^x$, $\tilde{S}^y$ and $\tilde{S}^z$ or the Pauli spin matrices (in the case of a qubit) do not commute. The inequality, $(\sigma^+)^2 + (\sigma^-)^2 + (\sigma_z)^2 \leq 1$, where the average is taken over an arbitrary state, suggests that for a mixed state the spin lies inside the Bloch sphere, whereas for a pure state it will lie on the Bloch sphere. In the latter case, the equality relation holds as in such a case it is a pure state. For a particle with spin $S$, this inequality reads $\langle S^x \rangle^2 + \langle S^y \rangle^2 + \langle S^z \rangle^2 \leq S^2$. If we consider $\langle (S^x)^2 \rangle + \langle (S^y)^2 \rangle + \langle (S^z)^2 \rangle$, the value will always be more than $S^2$ and will in fact be $S(S+1)$. This extra value comes from the fact that $\tilde{S}^x$, $\tilde{S}^y$ and $\tilde{S}^z$ do not commute. Thus, there is an uncertainty in determining the values of all three components of spin simultaneously. For a composite system, this results in extra correlations which essentially are non-local. For such a composite system, entanglement plays a role analogous to that of a one-qubit coherently superposed pure state that lies in the Bloch sphere. Since the susceptibility $\chi \propto (\langle M^2 \rangle - \langle M \rangle^2)$, the second term being less than or equal to $N^2S^2$, $N$ being the total number of spins, and the first term being proportional to $N^2S(S+1)$, the inequality given in equation (6) of Wieśniak et al [8] holds. Tóth [38] has discussed similar inequalities for entanglement detection in optical lattice of bosonic atoms with collective measurements.

We have analysed our magnetization and susceptibility data extensively and tried to ascertain the experimental validity of this relation. What follows is a careful exposition of the methodology and the validity of susceptibility measurements at high magnetic fields. Thereafter, we use the analysed experimental data to construct the $P$ and $Q$ variables mentioned after equation (15) and subsequently we test the $P$ and $Q$ inequalities and capture the QPT in copper nitrate at the lowest measurable temperature in our system.

The magnetization isotherm measured in the field range 0–7 T at $T = 2$ K for copper nitrate is given in figure 5. Although the entire temperature range has been shown in figure 2, here we focus on the data set below the antiferromagnetic ordering temperature, where all the
interesting features occur. The data were fitted to both the dimer and the alternating dimer models. Equation (16) gives the expression for a dimer model.

\[
M = \text{Tr}(\rho \cdot S_{\text{total}}^z) = \frac{2 \sinh(2g \mu_B H/k_B T)}{1 + 2 \cosh(2g \mu_B H/k_B T) + \exp(J/k_B T)},
\]

where \(g\) is the Landé \(g\)-factor.

The fit to this expression is given in red. We can see that the dimer model is not a very good representation of the real spin system. The expression for the alternating dimer model was generated numerically and was subsequently used to fit the magnetization data. \(T = 2\) K is used in the generated expression and fitting was done in the field range 0–7 T (blue curve). The expression is not explicitly shown here owing to its cumbrous nature. We can see that it is a very good fit. The values of \(J_1\) and \(J_2\) are 4 and 1 K, respectively, as obtained from this fit and consistent with earlier reports that \(J_2 = 0.25J_1\). This only goes to show that Cu(NO\(_3\))\(_2\) \(\times\) 2.5H\(_2\)O is an alternating dimer system as is also evident from neutron scattering data [29].

Hereafter, we analyse the susceptibility data as a function of magnetic field, acquired at the minimum temperature (2 K), shown in figure 6. The susceptibility data show a downturn after 4 T. Here, it is imperative to discuss the nature of measurement of generalized susceptibility employed in the MPMS or other magnetic susceptibility measurement systems.

The zero field magnetic susceptibility is defined as \(\chi = (\partial M_z/\partial H)\). From the fluctuation dissipation theorem, the magnetic susceptibility is given by

\[
\chi = \frac{1}{kT} \Delta^2(M_z) = \frac{1}{kT}(\langle M_z^2 \rangle - \langle M_z \rangle^2) = \frac{1}{kT} \left( \sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle - \left( \sum_{i=1}^{N} S_i^z \right)^2 \right),
\]

where \(N\) is the number of spins. The expression is given in equation (17).


Figure 6. Experimental data of magnetic susceptibility collected at \( T = 2 \) K for Cu(NO\(_3\))\(_2\) \( \times 2.5\)H\(_2\)O (open circles) as a function of magnetic field and the fit to the theoretical curve (solid line) represented by equation (18). The dotted line is a guide to the experimental data.

where \( \Delta^2(M_z) \) is the variance of magnetization. This definition of susceptibility gives a measure of the correlation function, \( \langle S_i^z S_j^z \rangle \), which could have a magnetic field dependence and one can measure the field dependence of susceptibility and find the field dependence of correlation function. This holds good till the magnetic field \( B \) is less than or of the order of exchange coupling \( J \). The approximation mentioned above breaks down the QCP, where \( B > J / k_B \) and the assumption of isotropy of space breaks down. In addition to the isotropy of space, there are other subtleties involved in the extraction of relevant quantum information from the susceptibility data as indicated below.

We have obtained a theoretical relationship for the field dependence of the correlation function and hence the susceptibility,

\[
\chi(H) = (g^2 \mu_B^2 N \beta) \left( 0.75 + \frac{1}{4} e^{-\left( \frac{J}{4} + H \right) \beta} + \frac{1}{2} e^{-\frac{J}{4} \beta} - \frac{3}{2} e^{-\frac{J}{4} \beta} + \frac{1}{4} e^{-\left( \frac{J}{4} - H \right) \beta} \right),
\]

where \( \beta = \frac{1}{k_B T} \).

The expression given in equation (18) is a generalization of equation (17) and was derived using the full Hamiltonian including the Zeeman or the field-dependent term. In deriving this we used the expression for expectation value in quantum statistical mechanics, namely, \( \langle \vec{S}_1 \cdot \vec{S}_2 \rangle = \text{Tr}(\exp(-\beta H) \vec{S}_1 \cdot \vec{S}_2) \), where \( H \) is the Hamiltonian for the alternating dimer model. Figure 6 shows the fit to the experimental susceptibility data as a function of field, collected at 2 K. Here, we have shown the plots only up to 4 T. The experimental value of susceptibility as a function of field (open circles) matches well with the theoretical value (bold curve). However, above the quantum critical field, there is a downturn in the data (see figure 6), as the experimentally measured susceptibility in the MPMS (which measures the derivative...
of magnetization) starts deviating from the generalized susceptibility above the QPT. This is because the magnetization saturates above the critical field and its derivative with respect to field will gradually go to zero. This will cause a reduction in susceptibility above the critical field that would eventually go to zero. This is reflected in the experimental susceptibility data (taken at 2 K) shown in figure 7, where we see a deviation in the theoretical fit from the experimental curve. Thus, in the experimental technique employed here to measure the susceptibility, one does not measure the magnetic field dependence of the correlation function, \( \sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle \), above the critical field. In this limit, i.e. above the QPT, these two are not the same, \( \chi(H) = \frac{1}{kT} \sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle - (\sum_{i=1}^{N} S_i^z)^2 \neq \partial M_z / \partial H \). The susceptibility measurement measures only the latter and this was also verified by taking the derivative of the magnetization data (\( \partial M_z / \partial H \)). The only way to ensure that one is really measuring \( \chi(H) = \frac{1}{kT} \sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle \) and not the derivative \( \partial M_z / \partial H \) is to measure the correlation function using inelastic neutron scattering measurements.

Another important point is that in a dimer-like antiferromagnetic system, since the average spin \( \langle S_i^z \rangle = 0 \), at least below a critical field, the ground state is the singlet Bell state \( |\Psi^-\rangle \). However, above a critical field where the QPT takes place, the ground state crosses over to a triplet-like state, which is a product state and \( \langle S_i^z \rangle \neq 0 \). In the definition of correlation function, \( (\sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle - (\sum_{i=1}^{N} S_i^z)^2) \), the second term will start contributing to the susceptibility, after the QCP. Thus, when we use the term \( \sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle - (\sum_{i=1}^{N} S_i^z)^2 \) in the susceptibility to plot \( P + Q \) as a function of field and temperature, it shows an asymmetry on either side of the QCP even at the lowest temperature. This is shown in figure 7(a), which gives the theoretical 3D plot of \( P + Q \) as a function of field and temperature. This is what one would obtain if we consider the experimental susceptibility in constructing \( Q \equiv 1 - \frac{kT}{N\chi} \). This asymmetry is in disagreement with what Wieśniak et al [8] obtained. If we ignore the second term in constructing susceptibility, i.e. if we consider only \( \sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle \), then we obtain the 3D ‘\( P + Q \)’ plot as obtained by Wieśniak et al and shown in figure 7(b). Figure 7(c) shows the side view of figure 7(b) to signify the dip in ‘\( P + Q \)’ around the critical field.

Figure 7. (a) Plot of theoretical \( P + Q \) as a function of magnetic field and temperature using \( (\sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle - (\sum_{i=1}^{N} S_i^z)^2) \) for constructing the susceptibility. (b) The same where susceptibility was constructed using \( \sum_{i,j=1}^{N} \langle S_i^z S_j^z \rangle \). (c) The side view of the theoretical \( P + Q \) (figure 7(b)) as a function of magnetic field and temperature.
Figure 8. Theoretical susceptibility as a function of field at temperature $T = 0.5\text{ K}$ using the correlation function $(\sum_{i,j=1}^{N} (S^z_i S^z_j) - (\sum_{i=1}^{N} S^z_i)^2)$ (+). The term $(\sum_{i=1}^{N} S^z_i)^2$ at $T = 0.5\text{ K}$ is shown in the figure (dotted curve). The curve ‘∗’ shows the susceptibility as a function of field at $T = 0.5\text{ K}$, obtained using $(\sum_{i,j=1}^{N} (S^z_i S^z_j))$. This is also obtained by adding the first two curves. The curve with the symbol ‘o’ represents $P$ and ‘x’ represents ‘$Q$’ at $T = 2\text{ K}$, where the latter uses the susceptibility plotted in the curve ‘∗’.

Before constructing the two-dimensional (2D) ‘$P + Q$’ as a function of temperature, one needs to demonstrate the different 2D curves of susceptibility mentioned above as a function of temperature. Figure 8 shows the susceptibility as a function of field at $T = 0.5\text{ K}$, constructed by using the correlation function $(\sum_{i,j=1}^{N} (S^z_i S^z_j) - (\sum_{i=1}^{N} S^z_i)^2)$ (curve ‘+’). The term $(\sum_{i=1}^{N} S^z_i)^2$ is also plotted (dotted curve) at the same temperature as a function of field. The sum of these two curves is also shown in this figure (curve ‘∗’). Next, we show the non-local quantity ‘$Q$’ as a function of magnetic field at $T = 0.5\text{ K}$ (curve ‘x’). This was constructed using the modified susceptibility (i.e. using the curve ‘∗’ mentioned above). The local term ‘$P$’ is also plotted in this figure (curve ‘o’) at the same temperature as a function of magnetic field. We have chosen $T = 0.5\text{ K}$ for all the 2D plots for consistency as we wanted to demonstrate the difference in the susceptibilities owing to different terms in the correlation function. This temperature was chosen to bring out the differences clearly as both ‘$P$’ and ‘$Q$’ are quite sharp at such a temperature.

Next, we show the theoretical plot of ‘$P + Q$’ as a function of magnetic field at $T = 2\text{ K}$ in figure 9(b). It shows a dip at the QCP, which is between 3.5 and 4 T. To match the experimental value of ‘$P + Q$’ with the theoretical value at $T = 2\text{ K}$, we consider the fitted experimental curves of magnetization and susceptibility shown in figures 5 and 6, respectively. Here, in constructing the experimental value of ‘$Q$’, we consider the susceptibility data up to the magnetic field where the theoretical curve matches the experimental data, i.e. up to the critical field. Beyond the critical field, we extrapolate the data up to 7 T using the parameters obtained from the fitted curve of figure 6. Since the experimental data measure the term $(\sum_{i=1}^{N} S^z_i)^2$ as well, we need to add this term to match the experimental data with the theoretical value of
Figure 9. (a) Plot of experimental ‘P + Q’ as a function of magnetic field at \( T = 2 \text{ K} \). Here the ‘Q’ used was obtained considering the corrected susceptibility. (b) Plot of theoretical ‘P + Q’ as a function of magnetic field at \( T = 2 \text{ K} \).

Wieśniak et al. This plot is shown in figure 9(a). The local property ‘P \( \equiv \frac{(\hat{M})^2}{N_b} \)’ was obtained entirely using the experimental magnetization data at \( T = 2 \text{ K} \).

One can clearly see the dip in the value of ‘P + Q’ in figure 9(a), corresponding to the QCP at around 3.5 T. This suggests that the ‘P + Q’ prescription proposed by Wieśniak et al indeed is a detector of QPT. Our experimental results also demonstrate the quantum information sharing and complementarity between the two observables, susceptibility and magnetization. The ‘P + Q’ inequality given in equation (15) is not violated in our system and the minimum value is indeed reached at QPT.

5. Conclusion

We have studied a Heisenberg spin chain compound known to exhibit dimer- or alternative dimer-like characteristics. The system has antiferromagnetic coupling and therefore exhibits an entangled ground state. This entanglement, which was extracted from the susceptibility data, decreases when the temperature is varied from the lowest attainable value (2 K) and vanishes around 5 K, where the antiferromagnetic correlations cease to exist. We also studied the magnetic field dependence of the magnetization and susceptibility and they both fit reasonably well to an alternating dimer model. We extracted the magnetic field dependence of entanglement and observed that the entanglement decreases with increase in field, owing to an increase in occupation of the excited states in comparison to the antiferromagnetic ground state. At the critical field, where there is a crossover between the ground state and the excited state, bringing about a change in symmetry of the ground states, a QPT occurs. Although we were successful in capturing the essential features of entanglement using a two qubit model, comparing it with the results obtained by Arnesen et al [16], we had to resort to an alternating dimer model to capture the QPT, which is a many-body effect. Translational invariance allows us to restrict ourselves to a finite-dimensional Hilbert space, instead of an infinite-dimensional Hilbert space.

An interesting possibility that follows from the fact that the system behaves like a chain of antiferromagnetically coupled spin half dimers, is that it is somewhat similar to a resonating...
valence bond like system \cite{39}. It separates into a set of dimers connected in a chain, such that the intra-dimer coupling is like a double bond, i.e. strongly connected, and the inter-dimer coupling is like a single bond, which is not so strong. However, a general term of this type could be claimed to evolve into another similar state where there is a role reversal of the different links (valence bonds) such that the single bond goes to double and vice versa and a new set of singlets are formed, displaced by a lattice site. The bonds jump to the adjacent site and back, such that one obtains a ‘resonating’ system. This symmetrization (symmetric superposition) of all possibilities is the usual way to obtain a quantum eigenstate of the lowest energy, assuming that the different terms are able to change into each other by a transition amplitude. At the QCP these dimers break and we have a new ground state. Thus, we see that in this picture also we need at least four spins that could be translated throughout the lattice to capture the whole system. Hence, an alternating dimer model comprising at least four spins can explain the QPT and also be able to capture the essential physics of entanglement and information sharing.

We have also explored the complementarity in the two observables, susceptibility and magnetization, which capture the non-locality and local nature of correlations in this spin system. Following an established prescription, the susceptibility and magnetization were suitably combined to form operators ‘$P$’ and ‘$Q$’, such that the minimum of the sum ‘$P + Q$’ when plotted as a function of magnetic field captures the QPT. From this measure, we could capture the QPT as a function of field at 2 K, showing that ‘$P + Q$’ is a good measure for detecting QPTs, even at non-zero temperatures. This could have significant implications in quantum information processing, as entanglement is known to scale close to QPT from theoretical results \cite{22, 23}. This scale invariance of entanglement coupled with the ‘$P + Q$’ measure could be harnessed to execute quantum communications in spin chains. In future, we are making an effort to further explore the QPT at lower temperatures. We also intend to do specific heat measurements to capture other aspects of entanglement and QPT in this system.

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