

Magnetic susceptibility as a macroscopic entanglement witness

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Abstract. We show that magnetic susceptibility can reveal spin entanglement between individual constituents of a solid, while magnetization describes their local properties. We then show that magnetization and its variance (equivalent to magnetic susceptibility for a wide class of systems) satisfy complementary relation in the quantum-mechanical sense. It describes sharing of (quantum) information in the solid between spin entanglement and local properties of its individual constituents. Magnetic susceptibility is shown to be a macroscopic (thermodynamical) spin entanglement witness that can be applied without complete knowledge of the specific model (Hamiltonian) of the solid.

Thermodynamical properties, such as heat capacity, magnetization or magnetic susceptibility, are normally ascribed to macroscopic objects with the number of individual constituent of the order of 10^{23} . In contrast, genuine quantum features like quantum superposition or entanglement are generally not seen beyond molecular scales. As mass, size, complexity and/or temperature of systems increase, the observability of their quantum effects is gradually limited by decoherence—an interaction of the system with its environment—that turns them into classical phenomena. This raises several questions: under which conditions can quantum features of individual constituents of a solid have an effect on its global properties? Can one detect existence

of quantum entanglement in a solid by observing its thermodynamical properties only? Can one consider macroscopic properties as quantum-mechanical observables in the sense that they obey complementary relations like position and momentum?

The complementarity principle is the assertion that there exist observables which are mutually exclusive in the sense that they cannot be precisely defined simultaneously. One of them, for example, the z component of the spin $\frac{1}{2}$ (σ_z), might be well defined at the expense of maximum uncertainty about the other orthogonal directions (σ_x and σ_y , σ 's are respective Pauli matrices). One can speak about sharing of (quantum) information between mutually complementary observables [1]. In the case of a qubit this can quantitatively be described by the relation $\langle \sigma_x \rangle^2 + \langle \sigma_y \rangle^2 + \langle \sigma_z \rangle^2 \leq 1$, where the average is taken over an arbitrary state. When extended to composite systems, the principle of complementarity asserts the mutual exclusiveness between entanglement and local properties of individual constituents of the composite system. In the case of two qubits this can be described by the relation $\sum_{i=x,y,z} \langle \sigma_i^1 \rangle^2 + \langle \sigma_i^2 \rangle^2 + \langle \sigma_i^1 \sigma_i^2 \rangle^2 \leq 3$, where the upper indices indicate qubits. The maximal value of 3 can be achieved, e.g., with product states (e.g. $\langle \sigma_z^1 \rangle = \langle \sigma_z^2 \rangle = \langle \sigma_z^1 \sigma_z^2 \rangle = 1$; others are zero) for which local properties of the qubits are well defined, but there is no entanglement. Alternatively, their joint properties could be well defined at the expense of a complete indefiniteness of the local properties (e.g. for a singlet state $\langle \sigma_x^1 \sigma_x^2 \rangle = \langle \sigma_y^1 \sigma_y^2 \rangle = \langle \sigma_z^1 \sigma_z^2 \rangle = -1$; others are zero).

Recently, a complementarity relation was proposed [2] between two macroscopic quantities, magnetization and magnetic susceptibility along *one* spatial direction. However, because entanglement between spin $\frac{1}{2}$ systems necessarily involves correlations at *different* spatial directions this cannot distinguish between classical and quantum correlations (not a motivation of [2]), which is the aim of the present paper.

Here we will show that magnetic susceptibility, when measured along three orthogonal spatial directions, can reveal entanglement between individual spins in a solid, while magnetization describes their local properties. We also show that for a large class of systems, magnetization and (zero-field) magnetic susceptibility, when combined in a particular way, satisfy a complementarity relation in the quantum-mechanical sense. This *macroscopic (thermodynamical) quantum complementarity relation* describes sharing of (quantum) information between entanglement and local properties of individual spins in the macroscopic solid sample (in an analogy with the relation given above for two qubits). To this end we will first prove that the sum of magnetic susceptibilities measured along x , y and z directions is a macroscopic witness of spin entanglement for a wide class of solid state systems. In contrast to internal energy [3]–[7], the present entanglement witness is more general (not only valid for special materials [8, 9]), can be directly measured in an experiment, and does not require the complete knowledge of the Hamiltonian of the system.

We consider a composite system consisting of N spins of an arbitrary spin length s in a lattice, which is described by a spin Hamiltonian H_0 . One should mention that entanglement in solids may exist in many different degrees of freedom, such as spin, spatial or occupation number degrees of freedom. In this paper we will refer to the spin, assuming, moreover, that spins can be precisely localized in sites of a lattice. Note, however, that other degrees of freedom could also sometimes be represented in the formalism of Pauli spin matrices [10]. Our results could, therefore, also be applicable to these other scenarios.

In order to study its magnetic response properties, the solid is now put in a weak magnetic field, say, directed along z -axis and of probe magnitude B_p resulting in an additional term $H_1 = B_p \sum_{i=1}^N s_z^i$ of the Hamiltonian (here and throughout the paper, the unit $\hbar = 1$ is assumed).

By s_a^i ($a = x, y, z$) we mean here an a th component of the i th spin operator in the lattice. Then the Hamiltonian becomes $H = H_0 + H_1$. When the system is in its thermal equilibrium under a certain temperature T , it is in a thermal state $\rho = e^{-H/kT}/Z$, where $Z = \text{Tr}(e^{-H/kT})$ is the partition function and k is the Boltzmann constant. From the partition function, one can derive all thermodynamical quantities, e.g. the magnetization $M_z = (1/Z\beta)(\partial Z/\partial B_p)$ or the magnetic susceptibility $\chi_z = (\partial M_z/\partial B_p)$, where $\beta = 1/kT$.

It can be shown that if $[H_0, H_1] = 0$, magnetic susceptibility is given by [11]:

$$\chi_z = \frac{1}{kT} \Delta^2(M_z) = \frac{1}{kT} (\langle M_z^2 \rangle - \langle M_z \rangle^2) = \frac{1}{kT} \left(\sum_{i,j=1}^N \langle s_z^i s_z^j \rangle - \left\langle \sum_{i=1}^N s_z^i \right\rangle^2 \right), \quad (1)$$

where $\Delta^2(M_z)$ is variance of the magnetization.

Microscopically, magnetic susceptibility is, in fact, a sum over all microscopic spin correlation functions $\langle s_z^i s_z^j \rangle$ for the sites i and j . The above is a very important relation as it connects a macroscopic quantity to its microscopic roots in the form of the two-site correlation functions. Note, however, that the nonzero value of the correlation function does not necessarily imply the existence of entanglement. What we need, loosely speaking, are sufficiently strong correlations in all three orthogonal spatial directions and they need to be combined in a specific way to reveal spin entanglement. This is the reason why we will now study the sum of magnetic susceptibilities χ_x , χ_y and χ_z for weak probe fields aligned along three orthogonal directions.

We now show that the expression $\chi_x + \chi_y + \chi_z$ is an entanglement witness. Entanglement witnesses in general are observables which (by our convention) have positive expectation values for separable states and negative ones for some specific entangled states [12]. The proof is based on the method of entanglement detection using the uncertainty relations [13]. For any separable state of N spins of length s (for any classical mixture of the products states, each appearing with probability w_n : $\rho = \sum_n w_n \rho_n^1 \otimes \rho_n^2 \otimes \dots \otimes \rho_n^N$), one has

$$\bar{\chi} \equiv \chi_x + \chi_y + \chi_z \geq \frac{Ns}{kT}. \quad (2)$$

We will now prove this inequality for product states of the spins and then the general result will follow directly due to convexity of separable states. Note that for an arbitrary state of spin s particle, one has $\langle (s_x)^2 \rangle + \langle (s_y)^2 \rangle + \langle (s_z)^2 \rangle = s(s+1)$ and $\langle s_x \rangle^2 + \langle s_y \rangle^2 + \langle s_z \rangle^2 \leq s^2$. If the thermal state was actually a product one of N spins, the variance of magnetization would be the sum of variances of individual spins: $\bar{\chi} = (1/kT)[\Delta^2(M_x) + \Delta^2(M_y) + \Delta^2(M_z)] = (N/kT) \sum_i [\Delta^2(s_x^i) + \Delta^2(s_y^i) + \Delta^2(s_z^i)] \geq N/kT(s(s+1) - s^2) = Ns/kT$. Note that this bound is also valid in the general case of separable states due to the convexity of the mixture:

$$\begin{aligned} \bar{\chi} &= (1/kT)[\Delta^2(M_x)_\rho + \Delta^2(M_y)_\rho + \Delta^2(M_z)_\rho] \\ &\geq (N/kT) \sum_n w_n \sum_i [\Delta^2(s_x^i)_n + \Delta^2(s_y^i)_n + \Delta^2(s_z^i)_n] \\ &\geq Ns/(kT), \end{aligned} \quad (3)$$

where index n denotes the n th subensemble in the mixture and $\Delta^2(X)_\rho$ is a variance of an observable X taken in a state ρ . The inequality (2) is saturated for any pure state which has a maximal magnetic quantum number m with respect to any direction (i.e. $|j, m = j\rangle$). The other

extreme case, of $\bar{\chi} = 0$ can be achieved for a singlet state of N spins where all three variances are equal to zero.

Therefore, if $\chi_x + \chi_y + \chi_z < Ns/kT$, the solid state system contains entanglement between individual spins. It is important to note that all susceptibilities should be taken for zero-fields B_p to ensure that they are measured for the same thermal state (for the same reason, no quantum phase transition at the points of the measurements is assumed). Because the measurement of the magnetic susceptibility has been an experimental routine for long time, it is clear that the present approach is an experimentally efficient method for detecting macroscopic spin entanglement. It might be of particular importance when there is only partial knowledge of the system's Hamiltonian and one thus has to rely more on experiment. In what follows, we will demonstrate the efficiency of the method using an exactly solvable model.

Suppose that the symmetry of the system is such that magnetic susceptibility is equal in all three directions $\chi_x = \chi_y = \chi_z$. This is the case, for example, for the Heisenberg spin lattices with isotropic, but in general inhomogeneous coupling constant J_{ij} : $H_0 = \sum_{i,j} J_{ij} \vec{s}_i \vec{s}_j$ (here the summation does not need to be constrained to nearest-neighbour interactions only). The entanglement criterion now reads as follows:

$$\chi_z < \frac{1}{3} \frac{Ns}{kT}. \quad (4)$$

We apply it to investigate the existence of entanglement in the infinite xxx Heisenberg chains of spins $\frac{1}{2}$ (figure 1(a)) and spins 1 (figure 1(b)), described by a Hamiltonian $H_0 = \sum_i \vec{s}_i \vec{s}_{i+1}$, at various temperatures. We use the results of [14] where the thermodynamic properties of the Heisenberg spin chains are obtained by the transfer-matrix renormalization-group method. The right-hand side of (4) is represented by the red solid lines in figure 1. The values of magnetic susceptibility left to the intersections point of the red and the theoretical curves cannot be explained without entanglement. The critical temperatures are $T_c = 1.6J$ for spins $\frac{1}{2}$ and $T_c = 2J$ for spins 1.

It is commonly believed that one reconstructs the physics of classical spins in the limit of infinitely large spins as suggested by the limit of the commutation relation for normalized spins $S_i \equiv \frac{s_i}{s}$ ($i = x, y, z$): $\lim_{s \rightarrow \infty} [S_x, S_y] = \lim_{s \rightarrow \infty} \frac{1}{s} S_z \rightarrow 0$. Our result shows, however, that the larger the spins are, the higher the critical temperature is below which entanglement is present. Perhaps, one possible explanation of this effect is that the longer spins produce entanglement of higher dimension than the one produced by shorter spins. Thus, for the same coupling strength J and temperature, the longer spins can develop larger amount of entanglement than the shorter spins, which might then persist at higher temperatures. The dependence of an amount of entanglement on dimensionality of subsystems was studied in [15].

It is interesting to compare the threshold temperature for the spin $\frac{1}{2}$ chain with the estimates based on other macroscopic entanglement witness such as internal energy. In [4, 16] it was shown that the internal energy can reveal the entire bipartite entanglement between neighbouring spins as measured by the concurrence [17]. Using these and the results from [14], we obtain that the concurrence vanishes below the threshold temperature of $0.795J$. The higher value of $1.6J$ as revealed by magnetic susceptibility can be explained by the fact that it, in contrast to internal energy, may also detect bipartite entanglement between non-neighbouring sites and multipartite entanglement.

We now turn to the derivation of a macroscopic quantum complementarity relation. We first note that the sum of the squares of magnetizations along three orthogonal directions satisfies the

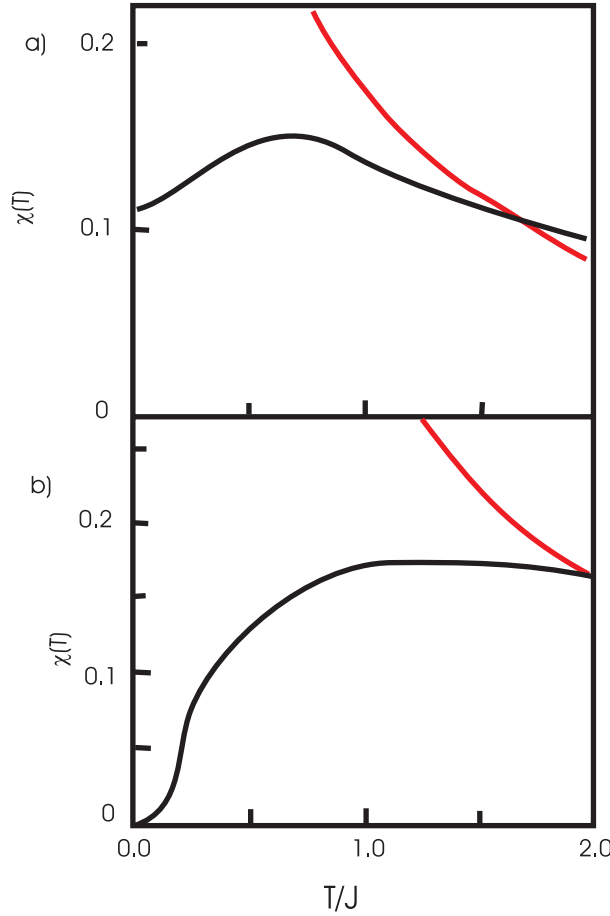


Figure 1. Detection of entanglement in the xxx Heisenberg spin $\frac{1}{2}$ (a) and spin 1 chains (b). The black solid curves are the theoretical curves from [14] and represent the temperature dependence of the zero-field magnetic susceptibility $\chi(T)$ per particle in the spin $\frac{1}{2}$ (a) and spin 1 chains (b). The red solid curves are from our work and represent the right-hand side of inequality (4). Mathematically, they represent hyperbolae $1/(6T)$ (a) and $1/(3T)$ (b). The critical temperatures below which entanglement exists in the chains are $T_c = 1.6J$ for spins $\frac{1}{2}$ and $T_c = 2J$ for spins 1 (the data in [14] were given only for $T \leq 2J$).

relation: $\langle \vec{M} \rangle^2 \equiv \langle M_x \rangle^2 + \langle M_y \rangle^2 + \langle M_z \rangle^2 \leq N^2 s^2$. This describes the complementarity between properties of individual spins in a solid, in analogy with the $\langle \sigma_x \rangle^2 + \langle \sigma_y \rangle^2 + \langle \sigma_z \rangle^2 \leq 1$ for a single qubit. If one of the observables in the sum, for example $\langle M_z \rangle^2$, takes its maximal value of $N^2 s^2$ (e.g. in state $|j = Ns, m = Ns\rangle$ where j is the angular and m the magnetic quantum number), the other two have to vanish. For the purposes of further discussion, we need the following relation between $\langle \vec{M} \rangle^2$ and $\langle \vec{M}^2 \rangle \equiv \langle M_x^2 \rangle + \langle M_y^2 \rangle + \langle M_z^2 \rangle$:

$$\langle \vec{M}^2 \rangle \geq \left(\frac{Ns+1}{Ns} \right) \langle \vec{M} \rangle^2. \quad (5)$$

Here follows the proof. Let us denote by $|j, m\rangle$ the joint eigenstates of \vec{M}^2 with eigenvalues $j(j+1)$ and M_z with eigenvalues m . Note that both sides of (5) are invariant under rotations

in the three-dimensional space. Thus, for any given state we can choose such a coordinate system that $\langle M_x \rangle = \langle M_y \rangle = 0$ and consequently $\langle \vec{M} \rangle^2 = \langle M_z \rangle^2$. Let us now define a new operator K such that $K|j, m\rangle = j|j, m\rangle$. Given that p_{jm} are probabilities of finding the system in any of the states $|j, m\rangle$, we have $\langle M_z \rangle = \sum_{j,m} p_{jm} m \leq \sum_{j,m} p_{jm} j = \langle K \rangle$. To complete the proof we use $\langle \vec{M}^2 \rangle = \langle K(K+1) \rangle$ and the fact that $Ns \geq j$ implies $Ns\langle K \rangle \geq \langle K^2 \rangle$. Thus we have

$$\langle \vec{M}^2 \rangle - \left(\frac{Ns+1}{Ns} \right) \langle \vec{M} \rangle^2 \geq \langle K(K+1) \rangle - \left(\frac{Ns+1}{Ns} \right) \langle K \rangle^2 \geq \left(\frac{Ns+1}{Ns} \right) \Delta^2(K) \geq 0.$$

We now exploit equations (1) and (5) to derive a macroscopic quantum complementarity relation:

$$\underbrace{1 - \frac{kT\bar{\chi}}{Ns}}_{\text{Non-local properties}} + \underbrace{\frac{\langle \vec{M} \rangle^2}{N^2 s^2}}_{\text{Local properties}} \leq 1. \quad (6)$$

The left-hand side of inequality (6) can be divided into two parts: $Q \equiv 1 - kT\bar{\chi}/Ns$ and $P \equiv \langle \vec{M} \rangle^2 / N^2 s^2$. While P describes the local properties of individual spins, Q is associated with quantum correlations between spins in a solid. This is because Q is proportional to two-site spin correlations for three orthogonal directions (three mutually non-commuting observables) and its positive value implies the existence of entanglement (see equation (2)). In the extreme case of a product state of N spins all aligned along the same direction (e.g. $|j=Ns, m=Ns\rangle$), their local properties are well defined ($P=1$) at the expense of no entanglement ($Q=0$). In the other extreme case, the state of the systems is highly entangled. Then non-local properties are maximally exhibited ($Q=1$), at the expense of $P=0$. In general, the relation (6) describes partial quantum information sharing between local and non-local properties of spins.

To illustrate the complementarity relation (6), we analyse a chain of antiferromagnetically coupled spin pairs—dimers—which are themselves uncoupled. This is a correct model for, e.g., copper nitrates and many organic radicals. The Hamiltonian in an external magnetic field of magnitude B is given by

$$H_0 = J \sum_j \vec{\sigma}^{2j} \cdot \vec{\sigma}^{2j+1} + B \sum_j \sigma_z^j. \quad (7)$$

The plot of P , Q and their sum $P+Q$ as a function of the magnetic field B and temperature T is given in figure 2. For $B=0$, the singlet is the ground state and the triplets are the degenerate excited states. For a higher value of B , however, the triplet states split and the gap between the singlet and first excited state $|-\rangle$ ($\sigma_z|-\rangle = -|-\rangle$) decreases. Therefore, in a thermal state at a given temperature as B is increased, the definiteness of non-local properties Q decreases because increasingly larger triplet component will be mixed with the singlet [18, 19]. On the other hand, as B increases the spins tend to orient themselves all parallel to the field, which results in higher values of magnetization and thus P , in agreement with the complementarity relation (see figure 2). Increasing T generally decreases $P+Q$ as thermal mixing has a destructive character both to Q and P . Note, however, that at all temperatures and all values of magnetic field the relation $P+Q \leq 1$ is satisfied.

In conclusion, we show that magnetic susceptibility is an entanglement witness for a wide class of systems. While magnetization describes local properties of individual constituents of a

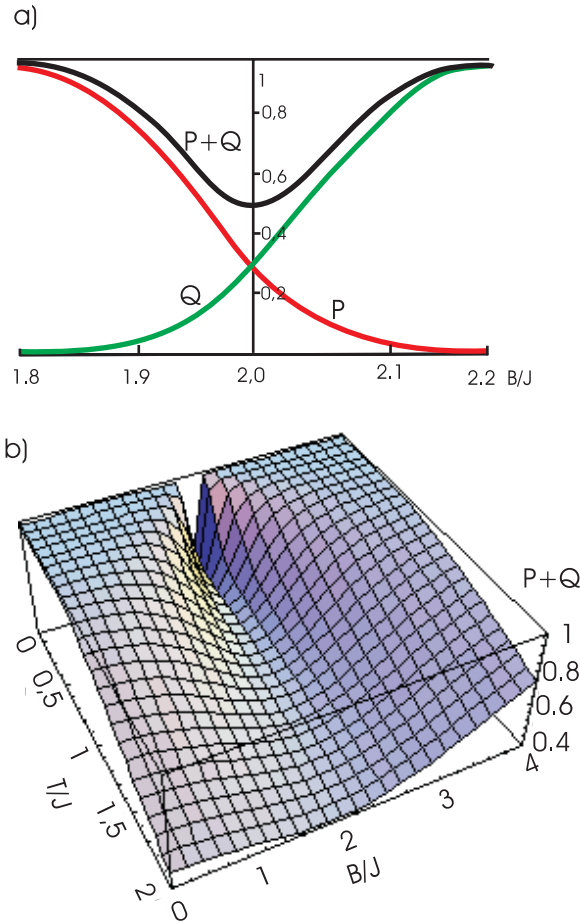


Figure 2. Macroscopic quantum complementarity relation between local (P) and non-local (Q) properties. (a) The plot of $Q = 1 - 2kT\bar{\chi}/N$ (red), $P = 4\langle \vec{M} \rangle^2 / N^2$ (green) and its sum $P + Q$ (black) for a chain of antiferromagnetically coupled spin $\frac{1}{2}$ pairs (dimers) versus the magnetic field B/J . The temperature is taken to be $T = 0.1J$. (b) The plot of $P + Q$ as a function of magnetic field B/J and temperature T/J . Under all temperatures and values of magnetic field, the complementarity relation $P + Q \leq 1$ is satisfied (see text for further discussion).

solid, the magnetic susceptibility specifies its spin entanglement. We show that magnetization and magnetic susceptibility satisfy a quantum complementarity relation. One of the quantities can thus increase only at the expense of a decrease in the other. This shows quantum information sharing in macroscopic quantum systems, such as solids. In the future, it will be interesting to investigate this feature at critical points where (quantum) phase transitions occur. It can be seen from our plot at $T = 0$ (figure 2(a)) that a sudden increase in non-local properties (Q) at the quantum phase transition point $B/J = 2$ must be accompanying with a corresponding sudden decrease in P . Otherwise, the complementarity relation would be violated.

Our results are not only relevant for fundamental research but also for quantum information science as they give the critical values of physical parameters (e.g. the high-temperature limit) above which one cannot harness quantum entanglement in condensed matter systems as a resource for quantum information processing.

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