Experimental observation of quantum entanglement in low-dimensional spin systems

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We report macroscopic magnetic measurements carried out in order to detect and characterize field-induced quantum entanglement in low-dimensional spin systems. We analyze the pyroborate $MgMnB_2O_5$ and the warwickite $MgTiOBO_3$, systems with spin 5/2 and 1/2, respectively. By using the magnetic susceptibility as an entanglement witness we are able to quantify entanglement as a function of temperature and magnetic field. In addition, we experimentally distinguish a system exhibiting a random singlet-phase state from one with the Griffiths phase. This analysis opens the possibility of a more detailed characterization of low-dimensional materials.

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Since the development of quantum mechanics, entanglement has been a subject of great interest. Lately, this is mainly due to the importance of entanglement in quantum information and computation. As a consequence, a great effort has been made to detect and quantify entanglement in quantum systems.[1] Quantum spin chains, a class of systems well known in solid-state physics, began to be studied in the framework of quantum information theory and there are proposals for the use of such systems in quantum computation.[2] Entanglement in interacting spin chains acquired relevance in the quantum information community, and therefore, there has been a special effort in understanding and quantifying quantum entanglement in solid-state systems.[3–7] At the same time, the condensed matter community began to notice that entanglement may play a crucial role in the properties of different materials.[8]

It is a difficult task to determine experimentally if a state is entangled or not. One of the new promising methods for entanglement detection is the use of entanglement witnesses (EW's), which are observables with negative expectation values for entangled states.[9,10] Magnetic susceptibility was recently proposed as an EW,[4] and some old experimental results were reanalyzed within this new framework.[11]

Entanglement appears in quantum spin chains, like the spin-1/2 Heisenberg model. The disordered spin-1/2 one-dimensional Heisenberg model, for example, presents a random singlet phase (RSP), where singlets of pairs of arbitrarily distant spins are formed.[12] Entanglement in spin chains has been quantified theoretically,[13] and a previous study showed its importance when characterizing a diluted magnetic material.[8]

Our investigation presents an experimental measurement of quantum entanglement in magnetic materials. As representative systems, we analyze the pyroborate $MgMnB_2O_5$[14,15] and the warwickite $MgTiOBO_3$,[16] two quasi-one-dimensional disordered spin compounds with previously known magnetic and thermodynamic properties that suggest the existence of either a RSP or a Griffiths phase[17] (GP) at low temperatures. Both systems present no thermal phase transition or spin glass freezing in the temperature range of the present study.

There are no experimental studies of random magnetic chains which discriminate between these two phases. In this paper, from a detailed analysis of magnetic measurements, we show unambiguously the existence of a RSP in $MgTiOBO_3$, which is expected for a spin-1/2 random exchange Heisenberg antiferromagnetic chain (REHAC). In addition, our study of $MgMnB_2O_5$ provides experimental evidence for the existence of a Griffiths phase in a low-dimensional system with spin $S>1/2$. Addressing the entanglement properties of these random systems, there is also a clear distinction between the RSP and the Griffiths phase. In the former, entanglement is well characterized and has been shown to scale with the logarithm of the size,[13,18] whereas in the latter there is no theoretical study of entanglement.

We make use of magnetization and ac susceptibility measurements as a function of temperature and applied magnetic field to detect and quantify entanglement by using the susceptibility as an entanglement witness.[4] First we show that both systems present entanglement at low temperatures with no applied field. Next, we analyze the ac susceptibility and magnetization as a function of field for different temperatures and we quantify the variation of entanglement as a function of applied field. Our analysis suggests that entanglement increases for increasing magnetic fields in a region of the *B*-*T* diagram. This unusual behavior was suggested by Arnesen *et al.*[3] and called magnetic entanglement.

In both pyroborate $MgMnB_2O_5$ and warwickite $MgTiOBO_3$ there are ribbons formed by oxygen octahedra sharing edges (see figures in Refs. 15 and 16, respectively). These octahedra give rise to four columns, along the ribbons, whose centers define a triangular lattice and two different crystallographic sites for metals: one in the central columns and another in the border ones. In the pyroborate such columns do not touch and both metal sites are equally occupied by the two metal ions.[19] In the warwickite, on the contrary, the columns do touch and the metal sites are probably occupied as in $MgScOBO_3$: 76% of the internal sites occupied by the transition metal and 24% by the alkaline-earth metal. The sites on the border columns have the opposite occupancy.[20] The nature of the electronic charge distribution and the superexchange interactions in the piroborates have been investigated in Ref. 21. These results show a clear admixture of the metal *d* orbitals with those of the ligand. This, together with some oxygen deficiency and that the interaction be-

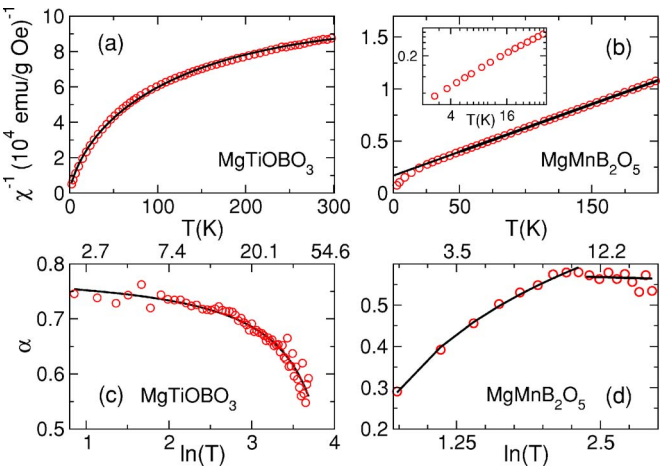

FIG. 1. (Color online) Upper: magnetic susceptibility versus temperature. (a) Experimental data for $MgTiOBO_3$ (open circles) and fitting using the susceptibility expression of a RSP (solid line). In panel (b), the experimental data for $MgMnB_2O_5$ (open circles) and for high temperatures a Curie-Weiss fitting (solid line). Lower: exponent α of $\chi \propto T^{-\alpha}$ as a function of ln T for (c) $MgTiOBO_3$ and (d) $MgMnB_2O_5$, the solid line in (c) is the theoretical curve for the exponent α for the RSP (see text).

tween octahedra extends beyond nearest neighbors, helps us to understand how positional disorder in the present systems leads to a distribution of exchange couplings.

The pyroborate powder was obtained from grinded single crystals, and the warwickite powder was directly obtained from its synthesis. The warwickite sample was analyzed through x-Ray diffractometry; it has been verified that the material was well crystallized and that its purity was 97.72%, as evaluated by the method of Lutterotti *et al.*[22] The more abundant impurity was the nonmagnetic $MgTiO_5$. More details on sample preparation were previously published.[15,16] Here dc magnetization and ac susceptibility measurements were performed with a commercial apparatus (Quantum Design PPMS).

In $MgTiOBO_3$, evidence for a RSP-like behavior was previously obtained from specific heat $C(T)$, susceptibility $\chi(T)$, and magnetization $m(H)$ measurements.[16] These quantities exhibit the characteristic power law behavior associated with a RSP, $\chi(T) \propto T^{-\alpha}$, down to the lowest measured temperature. In this system the magnetic ion Ti^{3+} has spin $S=1/2$, and due to the negligible magnetic anisotropy, this material is well described by a spin 1/2 (REHAC). The physical behavior is controlled by an infinite-randomness fixed point independent of the amount of disorder. On the other hand, in the $MgMnB_2O_5$ pyroborate, the magnetic ion Mn^{2+} is a spin-5/2, S-state ion. The phase diagram of a REHAC with $S \geqslant 1/2$ is not a trivial one. For weak disorder these systems present a GP, while only for strong disorder a RSP appears.[23]

In Figs. 1(a) and 1(b), we show the ac magnetic susceptibility as a function of temperature for $MgTiOBO_3$ and $MgMnB_2O_5$, respectively. Both systems have a sub-Curie regime at low temperatures. $MgMnB_2O_5$ acquires a Curie-like temperature dependence around 50 K. On the other hand, $MgTiOBO_3$ presents a sub-Curie susceptibility even at room temperature. It is known that both systems have a susceptibility which behaves as $\chi(T) \propto T^{-\alpha}$, although the temperature dependence of α was not further analyzed.[24]

These two different phases should be distinguished experimentally by the temperature dependence of the exponent α. The GP is characterized by a constant value of α.[25] For the RSP, we should expect a low-temperature susceptibility following $\chi(T) = \frac{1}{T \ln^2(\Omega_0/T)}$,[25] which is equivalent to $\alpha(T) = 1 - \frac{a}{\ln(T/\Omega_0)}$, where a is a constant.[26] So the RSP is characterized by a slowly varying $\alpha(T)$.

Following Hirsch,[26] we analyze the data by redefining $\alpha = -d(\ln(\chi))/d(\ln(T))$ and extract the temperature dependence of the exponent $\alpha(T)$ for both samples. Furthermore, we fit the experimental data of Fig. 1(a) using $1/\chi(T) = T \ln^2(\Omega_0/T)$ (solid line). Both Figs. 1(a) and 1(c) indicate that the susceptibility coincides exactly with the RSP model and α follows the same tendency previously predicted by numerical calculations.[26] In $MgMnB_2O_5$, previous assessments[15] and the inset of Fig. 1(b) suggest a power law behavior for $\chi(T)$ with a constant $\alpha \sim 0.55$. Within a more detailed analysis, shown in Fig. 1(d), we see an unequivocal slow increase of the exponent α, followed by a constant regime at intermediate temperatures. Although α is not constant in the whole temperature interval, as expected for a GP, its increase with T is clearly inconsistent with a phase governed by an infinite-randomness fixed point or RSP. However, for temperatures higher than 7 K, α is constant (up to 20 K) and this is consistent with the existence of a GP in this system.[27] The variation of α with temperature in $MgMnB_2O_5$ at low T may be related to a freezing of the Mn moments, as suggested by a maximum in the low-temperature susceptibility at $T_M \approx 0.6$ K with no corresponding anomaly in the specific heat.[14] Notice, however, that in the temperature range of the present experiments ($T > 1$ K) this system never enters in a spin glass state.[16]

We further investigate these two systems by comparing other independent measurements, such as magnetization and ac susceptibility as a function of a magnetic field, as shown in Fig. 2. From Figs. 2(a) and 2(b) we see that the $MgTiOBO_3$ data always present logarithmic corrections and the magnetization follows $M \propto \ln(B)$, as expected for a RSP.[25] On the other hand, for $MgMnB_2O_5$ both $\chi(B)$ and $M(B)$ follow a power law behavior with exponents $\alpha \sim 0.55$ and $1-\alpha \sim 0.45$, respectively. Such behavior is expected for systems in a GP.[27] Finally, for the $MgTiOBO_3$, we also analyze $\chi_{ac} \times T$ for different applied fields B [Fig. 2(c)]: the RSP is robust to applied fields below 3 T even at temperatures up to 100 K, where the susceptibility is not Curie like. However, the RSP characteristics disappear at high temperatures once the applied field is around 3 T with the appearance of a Curie-like behavior.

Once established that $MgTiOBO_3$ is in a RSP, we can expect the system to be entangled. Theoretically, the entanglement can be estimated by calculating the Von Neumann entanglement entropy of a subsystem A of the spin chain, with respect to a subsystem B. This quantity can be defined as $S = -\mathrm{Tr}\hat{\rho}_A \ln \hat{\rho}_A$. For a subsystem with length x embedded in an infinite system, the entanglement entropy for a Heisenberg chain in a RSP is given by $S = \frac{\ln(2)}{3} \ln(x)$, as

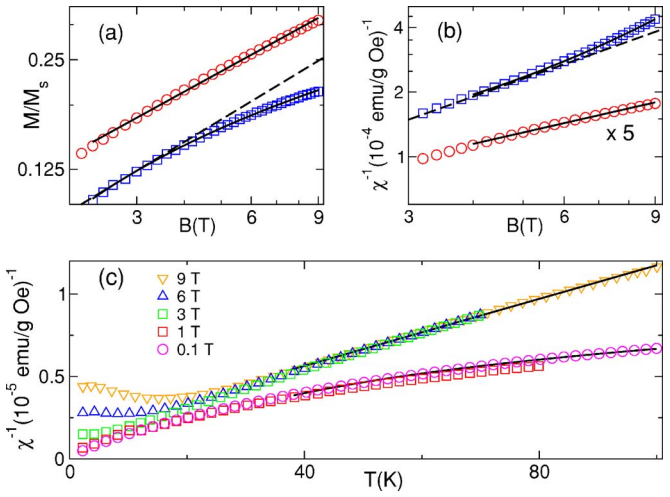

FIG. 2. (Color online) (a) Magnetization normalized by the saturation magnetization $M_s = g\mu_B Ns$ and (b) ac magnetic susceptibility as a function of the applied magnetic field for $MgTiOBO_3$ (open squares) and $MgMnB_2O_5$ (open circles), both for $T=2$ K. The solid lines represent a power law fitting for the $MgMnB_2O_5$ data and a logarithmic fitting for the $MgTiOBO_3$ data. (c) ac magnetic susceptibility ($H_{ac}=10$ Oe, $f=1$ kHz) as a function of temperature for different values of an applied dc field ($MgTiOBO_3$). The solid line indicates a Curie-law fitting at high temperatures.

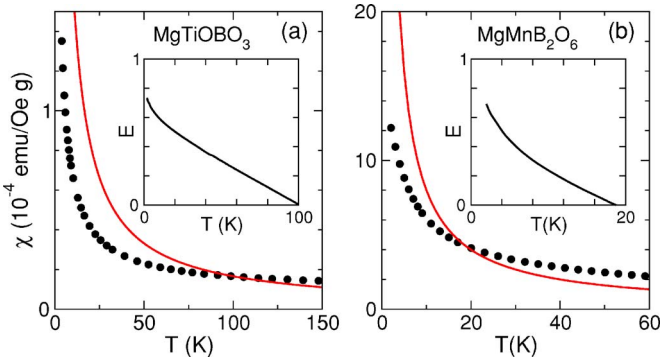

FIG. 3. (Color online) (a) Experimental data of magnetic susceptibility for $MgTiOBO_3$ (solid circles) and the limit for the EW (solid line). The system presents entanglement below the solid line. In panel (b) we show the same analysis for $MgMnB_2O_5$. The insets show the quantity E as defined in Eq. (1) which quantifies the entanglement detected by the EW as a function of temperature.

previously calculated by means of a RSRG[13] and further confirmed by numerical calculations.[18] From an experimental point of view, it is not possible to use the entanglement entropy to quantify the entanglement. Entanglement witnesses have been proposed and applied as an attempt to detect and quantify entanglement experimentally. The main advantage of EW's is that they are observables and their mean value can be directly measured. The use of magnetic susceptibility as an EW is based on the violation of local uncertainty relations.[28] Consider a set of Hermitian operators A_i. The uncertainty of an operator $\hat{A}_i$ for a given quantum state is defined as $\Delta A_i^2 = \langle A_i^2 \rangle - \langle A_i \rangle^2$, the statistical variance of the measurement outcomes. The uncertainty ΔA^2 can only be zero if the quantum state is an eigenstate of $\hat{A}_i$. A quantum state with zero uncertainty in all the properties $\hat{A}_i$ of the set must be a simultaneous eigenstate of these operators. If the simultaneous eigenstate does not exist, there must be a lower limit for the sum of the uncertainties. We can illustrate this concept for spins S where $s=(k-1)/2$ and $k \geqslant 1$ is an integer number: for any given spin state, we have $(\hat{S}_x^2 + \hat{S}_y^2 + \hat{S}_z^2)|\psi\rangle = s(s+1)|\psi\rangle$. On the other hand, the expectation values of $\hat{S}_i$ for $i=x,y,z$ define a vector with maximal length equal to $\pm s$. Using both relations, we obtain the inequality $\Delta S_x^2 + \Delta S_y^2 + \Delta S_z^2 = \langle \hat{S}_x^2 + \hat{S}_y^2 + \hat{S}_z^2 \rangle - (\langle \hat{S}_x \rangle^2 + \langle \hat{S}_y \rangle^2 + \langle \hat{S}_z \rangle^2) \geqslant s$.

We can apply this relation to obtain a limit for the correlation of separable states. Exemplifying for two spins 1 and 2: for product states, the uncertainty of $\hat{S}_i(1) + \hat{S}_i(2)$ is equal to the sum of the local uncertainties. On the other hand, maximally entangled states can have a total uncertainty of zero in all directions of $\hat{S}_i(1) + \hat{S}_i(2)$, *maximally violating the uncertainty relation*. In this case, the quantity $E = 1 - \sum_i \frac{\Delta[\hat{S}_{i=x,y,z}(1)+\hat{S}_i(2)]^2}{2s}$ measures the amount of entanglement verified by the violation of local uncertainties.[28,29] For a macroscopic system, we can generalize this quantity by using the magnetic susceptibility, which can be written as $\chi_i = \frac{1}{k_B T}\Delta^2 M_i = \frac{1}{k_B T}(\langle M_i^2 \rangle - \langle M_i \rangle^2)$, for $i=x,y,z$, where $\Delta^2 M_i$ is the statistical variance of the magnetization in the i direction.

Following Ref. 4, the entanglement can be measured by the quantity

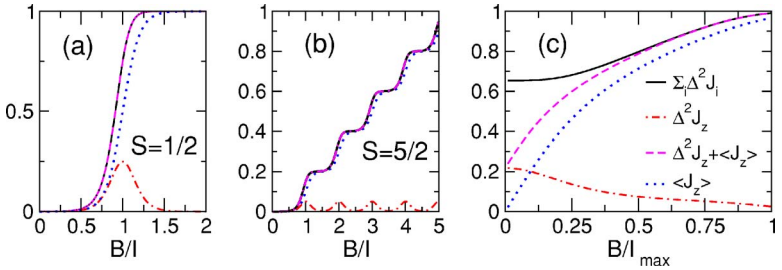

FIG. 4. (Color online) The sum of the total spin variance in three directions $i=x,y,z \Sigma_i \Delta^2 J_i$ (solid line), the spin variance in the z direction $\Delta^2 J_z$ (dot-dashed line), the sum $\Delta^2 J_z + \langle J_z \rangle$ (dashed line), and expectation value of the z component of the total spin $\langle J_z \rangle$ (dotted line) for (a) a pair of spins $S=1/2$, (b) a pair of spins $S=5/2$, and (c) a pair of spins $S=1/2$ interaction with a random interaction I_i (power law distribution and $T=0.05$) as a function of magnetic field. This is normalized by the exchange interaction and in the random case by the cutoff of the distribution.

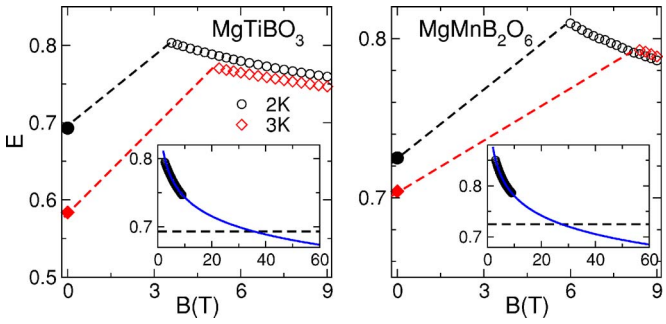

FIG. 5. (Color online) Experimental data for E using Eq. (3) for $B \neq 0$ for $MgTiOBO_3$ and $MgMnB_2O_5$. The insets show the extrapolation of E for very high values of B. The dashed lines are guides for the eyes for value of B where we do not have experimental results for E.

$$E = 1 - k_B T\left(\frac{\chi_x + \chi_y + \chi_z}{(g\mu_B)^2 Ns}\right). \tag{1}$$

In our case, s is the spin of a single magnetic ion (Mn or Ti) and N is the total number of spins per gram. First, we analyze a specific limit: if there is no dc applied field and the system is isotropic,

$$E = 1 - 3k_B T\chi_z/(g^2 {\mu_B}^2 Ns). \tag{2}$$

Since the studied samples have vanishing magnetic anisotropy, only one component of the susceptibility is needed to detect and quantify entanglement. For $\chi_z < \frac{g^2\mu_B^2 Ns}{3k_B T}$, the system is entangled and E quantifies the entanglement verified by the EW.

In Fig. 3 we show the experimental data for $MgTiOBO_3$ and $MgMnB_2O_5$: both systems present entanglement, although $MgTiOBO_3$ is entangled up to higher temperatures. The quantity E has a similar behavior as a function of temperature for both compounds, with a linear dependence on T for high temperatures.

If the system is subjected to an applied dc magnetic field, the ac susceptibility is no longer isotropic and Eq. (2) is not equivalent to Eq. (1). In order to study the entanglement in our systems under the effect of an external field without the need of transverse susceptibility measurements, we need to use the fact that both systems are in a phase where the spins form dimers.

For an applied dc field in the z direction, a pair of spins 1/2, where $\hat{J} = S(\hat{1}) + S(\hat{2})$, form a singlet $[\mathcal{H} = IS(\hat{1}) \cdot S(\hat{2})]$ at low fields. As $[\mathcal{H}, B\hat{J}_z] = 0$, the field does not modify the eigenstates, changing only their energies. At a given field, the energy of the singlet is no longer the lowest energy B_c, the singlet breaks, and the spins align with the field B. However, for the whole range of fields, the ground state of the system is such that spin variance is minimal: $\Delta J_x \Delta J_y = \frac{1}{2}\langle J_z \rangle$. In this case, as the system is isotropic in the x-y plane, we have $\Delta^2 J_y = \Delta^2 J_x = \frac{1}{2}\langle J_z \rangle$, so $\Delta^2 J_y + \Delta^2 J_x = \langle J_z \rangle$. This approximation is valid for $g\mu_B sB \gg k_B T$, which assures that other states, which do not have this property, are not populated. Similarly, the same approach holds for two pairs of spin 5/2 as shown in Figs. 4(a) and 4(b). As an illustration, we also consider a distribution of singlets, where the probability for interaction strength I follows a power law. As can be seen in panel (c) of the same figure, the approximation works well for high values of the magnetic field compared with the temperatures. Since both systems are in a phase where the spins form dimers ($MgTiOBO_3$ is in a RSP and $MgMnB_2O_5$ presents Griffiths singularities in a random dimer phase[30]), we can use this approximation to study quantum chains. It is possible to rewrite the EW as

$$E = 1 - \left(\frac{M_z}{g\mu_B Ns} + k_B T\frac{\chi_z}{(g\mu_B)^2 Ns}\right), \tag{3}$$

which does not make use of transverse susceptibility measurements but is valid only at high fields [as can be seen in Fig. 4(c)].

We perform the necessary measurements and using Eq. (1) for $B=0$ and Eq. (3) for high magnetic fields ($g\mu_B sB > 6k_B T$ for $MgMnB_2O_5$ and $g\mu_B sB > 2k_B T$ for $MgTiOBO_3$) we quantify the entanglement for both systems. In Fig. 5 we show that the magnetic field can increase entanglement in quantum spin chains, as theoretically suggested.[3,31] In the insets, we extrapolate the behavior of $E \times B$ for higher field values; measurements were performed with fields up to 9 T. From this extrapolation, we see that even if a field of 9 T is not high enough for the approximation made in Eq. (3), the extrapolation shows that the suggested increase of entanglement for low fields is still valid although the amount could be slightly overestimated.

In conclusion, by means of macroscopic magnetic measurements we fully characterize a random singlet phase in a low-dimensional spin system and it was possible to distinguish this phase from a Griffiths phase. We use an analysis where the magnetic susceptibility plays the role of an entanglement witness and measure the entanglement in two different spin systems as a function of temperature and magnetic field. The possibility of distinguishing experimentally between the RSP and GP gives additional motivation for the theoretical study of low-dimensional disordered quantum systems. Both types of analysis presented here can be used in conjunction to further characterize the phase diagram of these quantum systems.

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