Quantum discord in spin-cluster materials

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The total quantum correlation (discord) in Heisenberg dimers is expressed via the spin-spin correlation function, internal energy, specific heat or magnetic susceptibility. This allows one to indirectly measure the discord through neutron energy, specific heat or magnetic susceptibility. Using the available experimental data, we found the discord for a number of binuclear Heisenberg substances with both antiferro- and ferromagnetic interactions. For the dimerized antiferromagnet copper nitrate Cu(NO$_3$)$_2$ · 2.5H$_2$O, the three independent experimental methods named above lead to a discord of approximately 0.2–0.3 bit/dimer at a temperature of 4 K. We also determined the temperature behavior of discord for hydrated and anhydrous copper acetates, as well as for the ferromagnetic binuclear copper acetate complex [Cu$_2$L(OAc)] · 6H$_2$O, where L is a ligand.

PACS numbers: 03.65.Ud, 03.67.-a, 75.10.Jm, 75.50.Xx

I. INTRODUCTION

In the last years, it was understood that entanglement does not exhaust all quantum correlations in a system. Information correlations measured by entropy [1–3] were placed at the forefront. The total amount of correlations is identified with the mutual information $I$. The total correlations may be divided into two parts: purely classical correlations $C$ and purely quantum ones $Q$. The quantum excess of correlations, $Q = I − C$, has been called discord [4].

Note that the quantum discord has been evaluated explicitly only for several families of two-particle states (density matrices) [2,10].

It is remarkable that discord can exist even in separable (but mixed) states, i.e. when quantum entanglement is identically equal to zero. An example of two-qubit separable state with nonzero discord $Q = (3/4)\log_2(4/3) \approx 0.311$ was given in Ref. [11]. Below, in our paper, we will also discuss similar situations. As it turns out, “almost all quantum states have nonclassical correlations” [12]. Thus, quantum discord is a different measure of quantum correlation than entanglement.

As shown on the model of deterministic quantum computation with one pure qubit (DQC1) [11,13], quantum discord can lead to a speedup over classical computation even without containing much entanglement. Discord can also detect the quantum phase transitions [14,15]. Moreover, recently it has been shown [16] that in contrast to the entanglement and other thermodynamical quantities, discord makes it possible to identify the quantum phase transition points at finite temperatures ($T > 0$).

However, in order to utilize in practice the remarkable properties of quantum discord it is necessary to find methods to measure it experimentally. Our paper concerns this important problem. For the two-qubit Heisenberg systems, we found the relations between discord and the ordinary spin-spin correlation function, as well as between discord and internal energy, specific heat or magnetic susceptibility. This allows one to determine the behavior of quantum discord in various substances and investigate its properties.

The remaining part of this paper is organized as follows. In Section 2, we describe the model and present formulas for calculating the total, classical, and quantum correlations. Here a comparison analysis is also made for those correlations and the spin-spin correlation function and for entanglement. Section 3 is devoted to expressions of discord in terms of spin-spin correlation function, internal energy, specific heat, and magnetic susceptibility which are measured by standard experiments. These approaches are used on the copper nitrate compound. In this section we also present the temperature dependencies for the discord in crystals of copper(II) acetate complexes, which are excellent examples of dimeric materials. The results obtained are briefly summarized in Section 4.

II. CORRELATIONS IN HEISENBERG DIMER

The Hamiltonian of a Heisenberg dimer reads

$$\mathcal{H} = \frac{1}{2} J \vec{\sigma}_1 \cdot \vec{\sigma}_2,$$

(1)

where $J$ is the exchange coupling constant and $\vec{\sigma}_i = (\sigma^x_i, \sigma^y_i, \sigma^z_i)$ is the vector of Pauli matrices at the site $i = 1$ or 2. The magnetic moment components for the dimer are

$$M_\nu = \frac{1}{2} g_\nu \mu_B (\sigma^x_1 + \sigma^x_2), \quad \nu = x, y, z.$$

(2)

Here, $g_\nu$ are the components of the $g$ factor and $\mu_B$ is the Bohr magneton.

The density matrix of a system in thermal equilibrium has the Gibbs form

$$\rho = \frac{1}{Z} \exp(-\mathcal{H}/k_B T),$$

(3)
where \( k_B \) is the Boltzmann constant and \( Z \) the partition function which is found from the condition \( \text{Tr} \rho = 1 \). Performing necessary calculations for the system (1) one arrives at

\[
\rho(T) = \frac{1}{4} (1 + G \sigma_1 \sigma_2) = \frac{1}{4} \begin{pmatrix}
1 + G & 1 - G \\
2G & 1 - G \\
1 + G & 2G
\end{pmatrix}
\]

(4)

with

\[
G(T) = -1 + \frac{4}{3 + \exp(-2J/k_BT)}.
\]

(5)

It is easy to check that the quantity \( G \) equals the spin-spin correlation functions,

\[
G = \langle \sigma_1^x \sigma_2^x \rangle = \langle \sigma_1^y \sigma_2^y \rangle = \langle \sigma_1^z \sigma_2^z \rangle,
\]

(6)

where the brackets denote the statistical average. Values of \( G \) range from \(-1\) to zero for the antiferromagnetic cluster (\( J < 0 \)) and from zero to \( 1/3 \) for the ferromagnetic one (\( J > 0 \)).

The density matrix (4) has the form for which the entanglement of formation \([17–19]\).

Another type of quantum correlation in a system is the entanglement of formation

\[
E = -\frac{1 + \sqrt{1 - C^2}}{2} \log_2 \left( \frac{1 + \sqrt{1 - C^2}}{2} \right) - \frac{1 - \sqrt{1 - C^2}}{2} \log_2 \left( \frac{1 - \sqrt{1 - C^2}}{2} \right),
\]

(10)

where concurrence \( \tilde{C} \) for the ferromagnetic dimer completely vanishes, and for the antiferromagnetic one is \([20, 21]\).

\[
\tilde{C}(T) = \begin{cases} 
\frac{1}{2}(1 + 3G), & T < T_e; \\
0, & T \geq T_e.
\end{cases}
\]

(11)

Here the temperature \( T_e \) is given by equation

\[
k_B \frac{J}{T_e} = \frac{2}{\ln 3} = 1.8204 \ldots
\]

(12)

According to Eqs. (7)–(12), quantum discord and entanglement are functionally related.

In Fig. 1 the temperature dependencies of \( |G|, E, Q, \) and \( C \), which describe different correlations in the antiferromagnetic dimer are shown. (In an antiferromagnet, the correlator \( G \leq 0 \) therefore we took its absolute value.) It is seen that at \( T = 0 \) all correlations are maximal (saturated) and equal to one. The correlations preserve practically the same value with a small increase of temperature. This is associated with the existence of a gap in the energy spectrum of the discussed system. With a further increase in temperature, all kinds of correlations monotonically decrease. By this, the curves pass through the inflection points where a convexity is changed to concavity. Functions \( |G(T)|, Q(T), \) and \( C(T) \) are different from zero for all temperatures \( T < \infty \). When \( T \to \infty \), the value of spin-spin correlations tends to zero as \( |G| \sim 1/T \), and the information correlations \( Q(T) \) and \( C(T) \) go to zero more rapidly — according to the law \( 1/T^2 \).

From Fig. 1 it can be seen that at low temperatures the entanglement \( E \) is larger than both \( Q \) and \( C \). Because \( E(T) \) has the sudden disappear point at the temperature \( T_e \), the curve \( E(T) \) must intersect both \( Q(T) \) and \( C(T) \). Coordinates of intersection points are

\[
k_B T_{QE}/|J| = 0.5880 \ldots,
\]

\[
Q(T_{QE}) = E(T_{QE}) = 0.7462 \ldots \simeq 0.75
\]

(13)

and

\[
k_B T_{CE}/|J| = 0.9260 \ldots,
\]

\[
Q(T_{CE}) = E(T_{CE}) = 0.3390 \ldots
\]

(14)

The phenomenon that in some situations the entanglement can be larger that the total quantum correlations has been pointed out in Ref. [7]. From our calculations, it follows that by \( T \neq 0 \) discord is always greater than classical correlation, and the latter may be both larger and smaller than quantum entanglement.

At the temperature \( T_e \), entanglement in the antiferromagnetic dimer vanishes. On the other hand, the spin

FIG. 1: The temperature dependencies of correlations \(|G|, Q, C, \) and \( E \) for the antiferromagnetic Heisenberg dimer.
At zero temperature, $J > 0$ Eigenvalues of this matrix equal 1, and the total entropy correlations

$$I_e \equiv I(T_e) = 1 - \frac{1}{2} \log_2 3 = 0.2075 \ldots$$

and discord

$$Q_e \equiv Q(T_e) = \frac{1}{2} \log_3 \frac{2}{3} = 0.1258 \ldots$$

Consider now a dimer with the ferromagnetic coupling $J > 0$. At zero temperature, $G = 1/3$ and the density matrix (14) is

$$\rho_0 \equiv \rho(0) = \frac{1}{6} \begin{pmatrix} 2 & 1 & 1 \\ 1 & 1 & 2 \end{pmatrix} = \frac{1}{6} [2|00\rangle\langle 00| + |11\rangle\langle 11| + |01\rangle\langle 01| + |10\rangle\langle 10|].$$

Here $\{00, 01, 10, 11\}$ is the standard basis for the two-qubit system. The state (17) is mixed because $\rho_0^T \neq \rho_0$. At the same time, the state (17) is separable (i.e., the entanglement $E = 0$). Indeed, a partial transposition of $\rho_0$ is

$$\rho_0^T = \frac{1}{6} \begin{pmatrix} 2 & \ldots & 1 \\ \ldots & 1 & \ldots \\ 1 & \ldots & 2 \end{pmatrix}.$$

Eigenvalues of this matrix equal 1, 1, 1, and 3. All these eigenvalues are positive. Consequently, in accordance with the positive partial transpose (PPT) criterion (22, 23), the state (17) is separable. Using formulas (7–9) we find that the discord of a state (17) equals $Q_0 = 1/3 \simeq 0.333$. This value is larger than 0.311 for the discord of the example from Ref. [11] mention above.

Let us look at Fig. 3. The figure shows the dependencies of $G$, $Q$, and $C$ versus temperature for the dimer with the ferromagnetic coupling. Entanglement is absent in such a system. At zero temperature, the spin-spin correlation reaches its maximal value $G = 1/3$. Here quantum discord is also equal to 1/3. It is interesting that the total classical correlations are now less than $Q$ and according to (9) equal only to

$$C_0 \equiv C(0) = \frac{5}{3} - \log_2 3 = 0.0817 \ldots$$

Thus, the ratio of discord to classical correlation here achieves the value $Q_0/C_0 = 1/[5 - 3\log_2 3] \approx 4.0798$ (c.f. Ref. [24]).

At low temperatures all three types of correlations have quasi-horizontal sections (“pedestals”). With increasing temperature, the correlations pass through inflection points and then decay asymptotically to zero. Curves $G(T)$, $Q(T)$, and $C(T)$ do not at any point intersect with one another.

Notice the following. In accordance with Eqs. (7–9), the quantum discord $Q$ is a function of $G$ (see Fig. 3). This function is monotonic both for the antiferromagnetic and ferromagnetic dimers. Moreover, if $G = 0$ (absence of ordinary correlations) then also $Q = 0$ (absence of any quantum correlations), when $|G|$ is maximum ($= 1$ or $1/3$) then $Q$ takes the same maximum values. This allows us to consider spin-spin correlation as a measure of discord. This is similar to that of the concurrence as a measure of entanglement [25, 26].

III. EXPERIMENTAL MEASUREMENTS OF DISCORD

As mentioned above, discord has a number of attractive properties. Unfortunately, the question of how to measure it experimentally is open. But in the case of spin dimers, the information correlations $I$, $C$, and $Q$ can be expressed via the experimentally observed characteristics of a system.
A. Neutron scattering and discord

Inelastic scattering of thermal neutrons is a powerful tool for the study of low-energy excitations in crystalline transition-metal and other compounds. By this, the Fourier components of spin pair-correlation function are extracted from the scattering data ([27]–[29] and references therein). Performing the inverse Fourier transformation produces the correlation function itself.

The neutron scattering experimental results for the quasi-dimer antiferromagnetic crystals of deuterated copper(II) nitrate Cu(NO$_3$)$_2$·2.5D$_2$O are presented in Ref. [30] (see also [31]). (Notice that the deuteration needed for performing the neutron scattering experiments has no measurable effect on the exchange coupling [32].) In the experiments with the copper nitrate, the neutron scattering intensity was measured for the temperature range 0.31 < T < 7.66 K. The temperature $T_c$ is equal to about 5 K [31]. At $T = 4$ K, the spin-spin correlation has the value $G = -0.54(9)$ [33]. Using Eqs. (7)–(9) we estimate the discord as

$$Q = N_A Q_0$$

(as $2/0/./2.5D_2O$), where $c$ is the Avogadro number). However, we will normalize discord per a dimer.

B. Internal energy and specific heat

The internal energy per a mole of dimers [1] is given as

$$u(T) = -3RJ/2k_B G(T),$$

(20)

where $R = k_B N_A$ is the universal gas constant. In turn, the energy equals

$$u(T) = u_0 + \int_0^T c_m(T) dT,$$

(21)

where $c_m(T)$ is the magnetic part of specific heat (the part after subtraction of lattice contribution from the total heat capacity). The integration constant $u_0$ in Eq. (21) can be restored from the condition $u(\infty) = 0$, i.e.,

$$u_0 = u(0) = -\int_0^T c_m(T) dT.$$

(22)

Therefore, we can get the correlation function $G$, from calorimetric measurements, and then find the discord using the expression of $Q$ via $G$.

On the other hand, the magnetic specific heat per mole of dimers is given as [34]

$$c_m(T) = 12R \left( \frac{J}{k_B T} \right)^2 \frac{\exp(2J/k_B T)}{1 + 3 \exp(2J/k_B T)}.$$

(23)

FIG. 4: Relation of specific heat $c_m/R$ and correlation $G$. Dashed vertical line separates antiferro- and ferromagnetic regions. Points $G = -1$ and $G = 1/3$ correspond to the temperature $T = 0$ and $G = 0$ to the $T = \infty$.

This function exhibits a maximum (a Schottky-like anomaly). For the ferromagnetic coupling, $J > 0$, its coordinates are

$$k_B T_{\text{max}}/J = 0.9259 \ldots, \quad c_m^{\text{max}}/R = 0.1663 \ldots,$$

(24)

and for the antiferromagnetic one, $J < 0$,

$$k_B T_{\text{max}}/|J| = 0.7029 \ldots, \quad c_m^{\text{max}}/R = 1.0234 \ldots.$$  

(25)

Using Eq. (24) we can rewrite expression for the specific heat of Heisenberg dimers in the following forms:

$$c_m(T) = \frac{3}{4} \left( \frac{J}{k_B T} \right)^2 (1 + G(T))(1 - 3G(T))$$

(26)

and, especially remarkably,

$$c_m/R = \frac{3}{16} (1 + G)(1 - 3G) \ln^2 \left( \frac{1 + G}{1 - 3G} \right).$$

(27)

These relations can be used to extract the correlation $G$ directly from specific-heat measurements. The behavior of $c_m/R$ versus $G$ is shown in Fig. [4].

Consider as an example copper nitrate Cu(NO$_3$)$_2$·2.5H$_2$O. Its specific heat was measured for the temperature range 0.5 – 4.2 K [35] (see also [36] and references therein). At temperatures below 0.5 K, specific heat is zero. Near 1.82 K it has the maximum value of $2 \times 0.51R$. At higher temperatures, specific heat decreases obeying at $T \geq 4$ K the asymptotical law [35]

$$c_m/2R = 3.3/T^2.$$  

(28)

The value of the exchange constant has been estimated as $2J/k_B = -5.18$ K [35].

We have numerically integrated the available experimental data from zero to 4 K. Taking $u(0)/R = 3J/2k_B = -3.885$ K we determined that at $T = 4$ K
the internal energy is \( u(4)/R = -1.63 \text{ K} \) and hence \( G(4) \approx -0.42 \) [see Eq. (20)]. Consequently, according to the calorimetric data, the discord in the given binuclear cluster material at the temperature of 4 K equals \( Q = 0.19 \). This value can be considered to be reasonably in agreement with the estimate obtained from the neutron scattering measurements.

Note that the integration of function (28) from 4 K to infinity leads to \( u(4)/R = -1.65 \text{ K} \). Hence, \( G(4) \approx -0.42 \) and again return to the above result for the discord. On the other hand, according to Eq. (25) the specific heat at 4 K is \( c_m (4)/R = 0.4125 \). Solving the transcendental equation (27) and taking into account that the temperature 4 K is larger than the maximum temperature \( T_{\text{max}} = 1.82 \text{ K} \), we find \( G \approx -0.4 \) (see Fig. 4). Then \( Q \approx 0.18 \).

C. Magnetic susceptibility

Molar magnetic susceptibility of Heisenberg dimers satisfies the Bleany-Bowers equation [34, 37]

\[
\chi(T) = \frac{N_A g^2 \mu_B^2}{2 k_B T} (1 + G(T)).
\]

(29)

Here, \( g \) is the corresponding component of the Landé factor when the measurements are made on a single crystal or

\[
g^2 = \frac{1}{3} (g_x^2 + g_y^2 + g_z^2)
\]

(30)

if the measurements are performed on a polycrystalline (powdered) sample.

For the antiferromagnetic coupling \( J < 0 \), the Bleany-Bowers susceptibility displays a maximum with coordinates

\[
\frac{k_B T_{\text{max}}}{|J|} = \frac{2}{1 + W(3/e)} = 1.2472\ldots
\]

(31)

\[
\frac{|J| T_{\text{max}}}{N_A g^2 \mu_B^2} = \frac{1}{3} W(3/e) = 0.2011\ldots
\]

(32)

Here \( W(x) \) is the Lambert function defined by the equation \( We^W = x \). This function under the name LambertW(x) was included in the Maple package.

From Eq. (29), we get the spin correlation function

\[
G(T) = \frac{2 k_B T \chi(T)}{N_A g^2 \mu_B^2} - 1.
\]

(33)

That is

\[
G(T) = -1 + \frac{1}{2} \chi(T)/\chi_0(T),
\]

(34)

where

\[
\chi_0(T) = \frac{N_A g^2 \mu_B^2}{4 k_B T}
\]

(35)

is the Curie law for the paramagnetic ions. Hence, one can indirectly measure the discord by performing magnetometric measurements.

Note that the Bleany-Bowers susceptibility is related to the internal energy (20) as

\[
u(T) = -3 N_A J \left( \frac{k_B T}{N_A g^2 \mu_B^2} \chi(T) - \frac{1}{2} \right).
\]

(36)

Consequently, the magnetic specific heat is

\[
c_m(T) = \frac{3 k_B J}{g^2 \mu_B^2} \frac{\partial T \chi(T)}{\partial T}.
\]

(37)

Moreover, inserting (33) into equation (27) we obtain an expression for the specific heat \( c_m / R \) through the magnetic susceptibility without differentiation.

We analyze, in the third instance, the sample with the copper nitrate compound. The magnetic susceptibility of Cu(NO\(_3\))\(_2\)·2.5H\(_2\)O at low temperatures has been measured in Ref. [38]. The susceptibility of the powder has a rounded maximum at 3.2 K where it is equal to 2 × 0.065 emu/mol. For the model of binary clusters and isotropic exchange, the authors [38] found from the data that \( J/2k_B = 1.28 \text{ K} \) and \( g = 2.11 \). Using the experimental points presented in their figures (see Ref. [38]) and taking into account our normalizations per a mole of dimers, we found that at \( T = 4 \text{ K} \) the powder magnetic susceptibility is \( \chi = 2 \times 0.063 = 0.126 \text{ emu/mol} \). Then, Eq. (33) yields \( G(4) = -0.396 \). As a result, the discord is \( Q = 0.17 \). This estimate agrees more or less with the values obtained above from the neutron scattering data and heat capacity measurements. Some discrepancy can be attributed to experimental errors and especially to the fact that the copper nitrate is only quasi-dimeric.

We will now move on to other compounds, which belong to the pronounced binuclear materials.

1. Antiferromagnetic substances

Let us consider classical examples of spin-\( \frac{1}{2} \) Heisenberg dimeric materials — crystals of copper(II) acetates, \([\text{Cu(CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}]_2 \) (hydrated) and \([\text{Cu(CH}_3\text{COO})_2]_2 \) (anhydrous). Their magnetic susceptibility study has a long history beginning in 1915 and continuing into the present day [39–43]. Experimental results between 90 and 400 K are given in table form in [40]. The data have been described by the Bleany-Bowers equation with the fitted parameters \( 2J/k_B = -408 \text{ K} \), \( g = 2.13 \) for the copper(II) acetate monohydrate and \( 2J/k_B = -432 \text{ K} \), \( g = 2.17 \) for the anhydrous copper(II) acetate [40].

Using Eqs. (7–9), we obtained experimental points for the discord in both compounds. Results are presented in Fig. 5. Theoretical curves we plotted, taking the corresponding estimates for the coupling constants \( J/k_B \) and using Eqs. (9), (7–9).
We will now discuss the different domains in Fig. 5. According to Eq. (13) the points which are higher than the level \( Q \simeq 0.75 \) belong to the region where entanglement \( E > Q \). However, at temperatures \( T > 120−127 \) K, the discord becomes larger than the entanglement in both compounds. The temperatures of 371 K and 393 K that are marked on the abscissa axis by the longer bars are the points of sudden disappearance of entanglement in hydrated and anhydrous copper(II) acetates, respectively. At higher temperatures, the entanglement is zero. Near 400 K, the discord is \( 11−12\% \) of the maximum value \( Q(0) = 1 \).

2. Ferromagnetic compound

Entanglement in the ferromagnetic Heisenberg dimer is absent, but the discord is not zero.

We found in the recent literature the experimental data for the ferromagnetic dimer material — the binuclear copper(II) acetate complex \([\text{Cu}_2\text{L}(\text{OAc})] \cdot 6\text{H}_2\text{O}\), where \( \text{H}_3\text{L} = \text{2-(2-hydroxyphenyl)-1,3-bis[4-(2-hydroxyphenyl)-3-azabut-3-enyl]-1,3-imidazolidine} \). Magnetic susceptibility measurements for it have been performed between 5 and 300 K. Taking into account the crystal structure of this compound, the susceptibility data have been fitted to the Bleany-Bowers equation. The best least-squares fit has been obtained with the parameters \( J/k_B = 35.4 \) K and \( g = 2.13 \) [44]. At lowest measured temperatures near 5 K, the experimental points drop out from the theoretical dependence. This may be ascribed to the influence of weak interdimer couplings.

Using Eqs. (5), (7)–(9), and (34) we obtained from magnetic data the temperature behavior for the discord shown in Fig. 6. Maximal discord, \( Q \approx 0.32 \), is achieved near 13 K. At the temperature \( T = 300 \) K, the product \( \chi T = 0.89 \text{ cm}^3\text{K/mol} \) [44]. This leads to the discord \( Q \approx 0.003 \), i.e. about 1% relative to the theoretical limit of \( 1/3 \).

IV. CONCLUSIONS

We have related the quantum discord in binuclear spin clusters to the scattering data, as well as to the basic responses of a system to external perturbations — thermal and magnetic. This makes it possible to measure the discord in various magnetic substances.

In this paper, we have done a comparison analysis for the temperature behavior of discord, classical correlations, entanglement, and spin-spin correlation function in the two-qubit Heisenberg systems. We have shown that in the case of antiferromagnetic interactions, entanglement can be both larger and smaller than the discord or classical correlations. Discord and classical correlations can be present when entanglement is absent. This is observed both in antiferro- and ferromagnetic dimers.

We have presented the temperature dependencies of discord for different solid-state dimeric materials with antiferro- and ferromagnetic couplings.

In the current literature, the transfer of quantum correlations from one system to another is intensively discussed (e.g., [45–47] and references therein). In particular, several schemes for extracting the entanglement from solids have been reported. With respect to discord, its dynamical behavior under the quantum state transfer, for example, between semiconductor double-dot molecules and photons is being studied now [48].

ACKNOWLEDGMENT

I am grateful to E. I. Kuznetsova for her help in the work.

This research was partially supported by the programs Nos. 18 and 21 of the Presidium of RAS.
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