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On the way to creation of materials for quantum computers

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Abstract. General requirements for the materials for systems and devices whose operation is based on quantum correlations are discussed. The fluctuation theory of quantum entanglement is outlined. This theory allows one to estimate the size of the region of weak fluctuations of quantum entanglement for spin dimers in various materials. The concept of quantum discord as a measure of pure quantum correlations is outlined. Quantum correlations in polycrystalline samples of binuclear nitrosyl iron complexes were estimated from the temperature dependences of the magnetic susceptibilities. Particular attention is paid to calculations of the temperature corresponding to the emergence of quantum entanglement. Methods for increasing this temperature are discussed. The problem of the quantum information transmission as well as the future development of quantum technology is considered. The bibliography includes 120 references.

I. Introduction

The development of new technologies is immediately related to the use of such physical resources, as materials , various types of interactions and energy sources. In the 20th century, this list of technologically important resources was augmented with information that has become in great demand since the invention of computers that process complex information outside human brain. According to Moore's law, the performance of microprocessors is doubled every 18 months. This is mainly due to miniaturization of microprocessors and the time when logic elements in microchips will be made of a few atoms seems not to be far out. This will be followed by the need to take into account quantum effects.

Thus, the existing technologies should be replaced by a new quantum technology.[1] Its potential is much greater than that of the technology based on small fast microprocessors whose operation is based on the laws of classical physics. Calculations that allow one to solve complicated tasks inaccessible to any classical computer [2] are at the back of quantum technology. Based on the quantum technology, methods of quantum cryptography were developed, which make it possible to securely transmit information.[1, 2] This technology enables practical implementation of quantum teleportation,[1] quite a recent subject of Sci-Fi books, and opens great prospects for the development of quantum metrology [3 – 5] to elaborate new highly sensitive methods of measurements of physical parameters with high resolution.

Of course, quantum technology uses specific features of systems that obey the laws of quantum mechanics. Comparing the descriptions of classical and quantum systems, a basic difference between them should be emphasized. If a classical system is exactly defined, all subsystems of the system are also known. *Vice versa*, if all subsystems of a classical system are specified, the system is known exactly. For quantum mechanical systems, these rules are not always met. For instance, if a state of a quantum system is known, this does not mean exact knowledge of states of all subsystems of the system in question.[2] Such states of quantum systems are called entangled states.[2, 6, 7]

Quantum entanglement is one of the most intriguing properties of quantum mechanical systems. The concept of quantum entanglement was proposed for the first time by Einstein, Podolsky and Rosen.[8] It demonstrated a correla-

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tion of the states of two particles separated by a long distance. A change in the state of one particle led to a change in the state of the second particle although no interparticle interactions occurred. This *Gedankenekseperiment* was treated as proof of incompleteness of quantum theory.[8] However, more recently, the existence of quantum mechanical entangled states was proved experimentally.[9] Nowadays,[2] one can say with certainty that it is the existence of entangled states that is responsible for the main advantages of quantum devices (in particular, quantum computers).

Of course, quantum entanglement is different in different systems. To choose materials suitable for quantum device applications, a quantitative measure of quantum entanglement is required. Various measures of quantum entanglement were introduced.[10–13] Their calculations include a labour-consuming procedure of optimization over various parameters of the system. Hill and Wootters[11,12] succeeded in carrying out an analytical optimization procedure and deriving a simple expression for calculations of quantum entanglement in two-particle systems with the spins $s = 1/2$ (spin dimers). The Wootters relation[12] is widely used in studies of quantum entanglement in the materials containing dimeric systems.[14–25] It also allows one to solve the problem of a register of a quantum computer.[26–31] Indeed, exponential acceleration of the performance of a quantum computer compared to classical analogues is possible if all pairs of particles of the register are entangled and have nearly equal quantum entanglement values. Meeting this condition requires the choice of materials with rather strong interaction between adjacent particles. Also, it is significant that the time characterizing the interaction of the system under study with other degrees of freedom (decoherence time) be long enough. Practically, it is appropriate to operate quantum devices at relatively high temperatures, *i.e.*, the requirements listed above should be met at not too low temperatures.

Summing up, one should choose materials containing systems with (i) controllable quantum entanglement at not too low temperatures and (ii) long decoherence times. Quantum entanglement is defined through entropy, a fluctuating statistical quantity. Therefore, fluctuations of quantum entanglement are also possible.[32] This means that materials should be chosen taking into account not only the quantum entanglement, but also its fluctuations. An ideal material contains a system with a high quantum entanglement, small entanglement fluctuations and long decoherence time at a specified temperature.

However, recent studies revealed that in some cases, this scenario of the choice of materials for quantum devices is too cumbersome. It was found[33–35] that sometimes the quantum properties of materials are also manifested in the separable states. For instance, exponential acceleration of performance can also be attained with a register formed by a quantum system in a mixed non-entangled separable state.[36–38] This can be explained by the fact that 'quantumness' of the system is determined by the quantum correlations while entanglement characterizes only a proportion of these correlations. Ollivier and Zurek[39] showed that quantum correlations can be described using the quantum discord (difference between the total and purely classical correlations) or mutual quantum information.[40] The quantum discord can differ from zero for the states with zero quantum entanglement. Thus, quantum discord covers a larger class of quantum correlations compared to entanglement, thus being more efficient when choosing materials for quantum device applications.

In the present review, we will consider the concepts of quantum entanglement, quantum entanglement fluctuations and quantum discord. The results of experiments on measurements of quantum entanglement and quantum discord in various materials are described in detail. Based on these data, the requirements for materials to be used in quantum devices are discussed. Evaluation of quantum correlations is illustrated taking the magnetic susceptibility measurements for polycrystalline samples of two binuclear nitrosyl iron complexes as examples. For one complex, the temperature dependences of quantum entanglement, quantum entanglement fluctuations and quantum discord were studied. Methods of determination of the temperature corresponding to the onset of quantum entanglement (so-called entanglement temperature T_e), and methods for increasing T_e are discussed. The problem of quantum information transmission is discussed. Prospects for the development of quantum computing, quantum teleportation, quantum cryptography and quantum metrology are outlined.

II. Entangled states in quantum mechanics

Consider a system of two spins s ($s = 1/2$). Each spin is described by three Pauli matrices[41,42]

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad (1)$$

$$\sigma_y = \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix},$$

$$\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},$$

written in the basis corresponding to the diagonal matrix σ_z. It is convenient to write this basis in the Dirac notation,[41] *viz.*, $|0\rangle$ and $|1\rangle$ stand for the spin states with the spin projections on the z axis equal to $1/2$ and $-1/2$, respectively. An isolated quantum system is completely described by the wave function $|\Psi\rangle$, *i.e.*, is in a pure state.[42] Then, the spin is used for practical implementation of the simplest carrier of quantum information called a qubit (quantum generalization of a bit).

Consider two different systems with the wave functions

$$|\Psi_1\rangle = |00\rangle, \quad (2)$$

$$|\Psi_2\rangle = \frac{1}{\sqrt{2}}\Big(|00\rangle + |11\rangle\Big).$$

In the system with the wave function $|\Psi_1\rangle$, the projections of both spins are known. In the system with the wave function $|\Psi_2\rangle$, the situation is more complicated. On the one hand, both spin orientations in this system are unknown; on the other hand, having measured the orientation of one spin, the orientation of the other spin becomes known immediately. The state $|\Psi_2\rangle$ has no analogues in the classical physics. The quantum state described by the function $|\Psi_2\rangle$ is called entangled state[2,10,14,43–45] and the phenomenon of existence of such states is called quantum entanglement.[2]

Using the Dirac formalism,[41] the states (2) can be described by the density matrices

$$\rho_1 = |00\rangle\langle 00|, \tag{3}$$
$$\rho_2 = \frac{1}{2}\Big(|00\rangle + |11\rangle\Big)\Big(\langle 00| + \langle 11|\Big),$$

where ρ_1 and ρ_2 are the 4×4 Hermitian density matrices.[41] From this point on, we will call the first spin 'subsystem A' and the second spin 'subsystem B'. Averaging of the system with the density matrix ρ_1 over the states of the second spin (*i.e.*, calculation of the trace over states of the subsystem B) gives the so-called 2×2 reduced density matrix for the subsystem A

$$\rho_1^A = |0\rangle\langle 0|. \tag{4}$$

Analogous reduction in the system with the density matrix ρ_2 gives

$$\rho_2^A = \frac{1}{2}\Big(|0\rangle\langle 0| + |1\rangle\langle 1|\Big). \tag{5}$$

The density matrix ρ_1^A gives unambiguous orientation of the first spin, but the density matrix ρ_2^A only gives an equal probability of 1/2 for both possible orientations of the first spin. Therefore, the von Neumann entropy of the system with the density matrix ρ_1^A

$$S_1^A = -\mathrm{Tr}\left\{\rho_1^A \log_2 \rho_1^A\right\} \tag{6}$$

equals zero and the entropy of the system with the reduced density matrix ρ_2^A

$$S_2^A = -\mathrm{Tr}\left\{\rho_2^A \log_2 \rho_2^A\right\} \tag{7}$$

is unity.

It is significant that reduction over states of the first spin leads to the same result.[10] Thus, the entropy determined using the reduced density matrix of two-particle system is independent of the choice of the subsystem and can be considered as a quantitative characteristic (measure) of quantum entanglement (E) in this system. If this entropy is equal to zero, one deals with a zero-entanglement system.[10]

Thus, the entropy of the reduced state can be used as a measure of quantum entanglement in a pure state of a two-qubit system[46] when the system is described by the wave function and the density matrix meets the 'purity' criterion[10]

$$\rho = \rho^2. \tag{8}$$

One can check with ease that the density matrices ρ_1 and ρ_2 meet criterion (8).

The entropy of a binary system in a mixed state,[10] when condition (8) is not met, cannot be used as a measure of quantum entanglement. Such binary systems are described by density matrices rather than wave functions. Taking a set of pure states $|\Psi_i\rangle$ such that the density matrix of a binary system (each subsystem of the system contains an arbitrary number of qubits) can be represented in the form

$$\rho = \sum_i p_i |\Psi_i\rangle\langle\Psi_i|\,, \tag{9}$$

where $p_i \geqslant 0$ are the weighting factors

$$\sum_i p_i = 1,$$

the measure of quantum entanglement $E(\rho)$ in the mixed system can be defined as[10]

$$E(\rho) = \min_E \sum_i p_i S(\Psi_i), \tag{10}$$

where $S(\Psi_i)$ is the entropy of the reduced pure state $|\Psi_i\rangle$ and the minimum is taken over the ensemble $E(\rho) = \{p_i, |\Psi_i\rangle\}$ which defines the density matrix (9).

The definition of quantum entanglement using relation (10) even for a two-particle system is a labour-consuming process because it involves the optimization procedure. Therefore, considerable efforts were made to simplify expression (10). Hill and Wootters[11,12] succeeded in doing so only for two-qubit systems.

To clarify the main idea of Wootters,[12] it is convenient to assume that a system with the wave function $|\Psi_1\rangle$ [see expression (2)] includes two spins oriented along some z axis in the spin space. By inverting both spins using the Pauli matrix σ_y, *i.e.*, by transforming $|\Psi_1\rangle$ as follows:

$$|\widetilde{\Psi}_1\rangle = (\sigma_y \otimes \sigma_y)|\Psi_1\rangle, \tag{11}$$

($\otimes$ means the tensor product),[47] one gets the function $|\widetilde{\Psi}_1\rangle$ orthogonal to $|\Psi_1\rangle$

$$\langle\widetilde{\Psi}_1|\Psi_1\rangle = 0.$$

Having done an analogous transformation with the function of the maximally entangled state $|\Psi_2\rangle$, one gets

$$|\langle\widetilde{\Psi}_2|\Psi_2\rangle| = 1.$$

Thus, the quantity

$$C = |\langle\Psi|\widetilde{\Psi}\rangle| \tag{12}$$

allows one to distinguish between entangled and non-entangled (separable) states. This quantity is called concurrence.[11] It was found that in the general case of a mixed two-qubit system the entanglement $E(\rho)$ is a function of concurrence

$$E(\rho) = H(C(\rho)), \tag{13}$$

where

$$H(C) = H\left(\frac{1+\sqrt{1-C^2}}{2}\right) \tag{14}$$

and $H(x)$ is the Shannon function[48]

$$H(x) = -x\log_2 x - (1-x)\log_2(1-x). \tag{15}$$

The function $H(C)$ monotonically increases from 0 to 1 as the concurrence increases from 0 to 1. For this reason, the concurrence can be used as a measure of quantum entanglement.

Wootters showed[12] that

$$C(\rho) = \max\{0, \lambda_1 - \lambda_2 - \lambda_3 - \lambda_4\}. \tag{16}$$

Here $\lambda_1 \geqslant \lambda_2 \geqslant \lambda_3 \geqslant \lambda_4$ are the eigenvalues of the matrix

$$R = \sqrt{\sqrt{\rho}\,\widetilde{\rho}\sqrt{\rho}},$$

where

$$\widetilde{\rho} = (\sigma_y \otimes \sigma_y)\rho^*(\sigma_y \otimes \sigma_y). \tag{17}$$

Complex conjugation (ρ^*) is done in a standard basis, which for two spins $s = 1/2$ has the form $|\uparrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$, $|\downarrow\downarrow\rangle$. Relations (13)–(17) allow one to calculate the quantum entanglement of an arbitrary state of the two-qubit system.

Other measures of quantum entanglement in quantum mechanical systems have also been proposed.[44,45] Among them, there is the so-called double negativity based on the positive partial transpose (PPT) procedure. This measure is efficient for estimation of bipartite quantum entanglement between two multiparticle subsystems.[13,49,50] To clarify the essence of this measure, consider a quantum mechanical system S and two subsystems, A and B, of S. Let subsystem C include all remaining particles of the system, so that

$$S = A \cup B \cup C.$$

The double negativity is the double sum of the moduli of all negative eigenvalues of the matrix $(\mathrm{Tr}_C\rho)^{\mathrm{T}_A}$, the trace being calculated over the subsystem C; the upperscript T_A denotes transposition over the subsystem A.

It should be noted that if A and B are single-qubit subsystems, it is possible to use both measures (Wootters measure and double negativity) and both of them give equivalent results (equal results for the two-particle system). The main value of the double negativity consists in that it can be used in practice for subsystems containing any number of particles, whereas the Wootters relation is only applicable to one-qubit subsystems.

III. Fluctuations of quantum entanglement

In the preceding Section we demonstrated that quantum entanglement is related to entropy. However, entropy is a statistical quantity and can fluctuate.[46] Therefore, quantum entanglement fluctuations should also be taken into account. Of course, for a large set of statistically independent subsystems the volume-averaged relative fluctuations of physical properties are negligible,[46] but for single composed systems the quantum entanglement fluctuations can be significant and even reach infinitely large values.[32]

Now we will return to the definition of quantum entanglement in a two-particle system comprising two subsystems A and B in a pure state Ψ. The quantum entanglement in this system is determined[11] by the von Neumann entropy of any subsystem

$$E = \overline{S}(\rho_A) = \overline{S}(\rho_B) = -\mathrm{Tr}\{\rho_A \log_2 \rho_A\} = -\mathrm{Tr}\{\rho_B \log_2 \rho_B\}, \tag{18}$$

where

$$\rho_A = \mathrm{Tr}_B|\Psi\rangle\langle\Psi|,$$
$$\rho_B = \mathrm{Tr}_A|\Psi\rangle\langle\Psi|$$

are the reduced density matrices of the subsystems A and B, respectively.

Reduction involves the averaging of the density matrix over one subsystem (*e.g.,* over states of one spin). However, in fact one subsystem 'senses' the state of the other subsystem involved in the averaging procedure; as a result, the entropy of quantum entanglement appears to be a flutuating quantity.[32] Following the general definition, the root-mean-square fluctuation of a random variable (ΔE) is[46]

$$\Delta E = \left[\overline{S}^2 - (\overline{S})^2\right]^{1/2}, \tag{19}$$

where the first and second moments $\overline{S}$ and $\overline{S}^2$, respectively, are taken for one subsystem, A or B.

Consider the behaviour of quantum entanglement fluctuations in the two-particle (two-qubit) model.[32] The wave function of the model written in the standard basis has the form

$$|\Psi\rangle = a|00\rangle + b|01\rangle + c|10\rangle + d|11\rangle, \tag{20}$$

where

$$|a|^2 + |b|^2 + |c|^2 + |d|^2 = 1.$$

The reduced density matrices ρ_A and ρ_B are as follows:

$$\rho_A = \begin{pmatrix} |a|^2 + |b|^2 & ac^* + bd^* \\ a^*c + b^*d & |c|^2 + |d|^2 \end{pmatrix}, \tag{21}$$

$$\rho_B = \begin{pmatrix} |a|^2 + |c|^2 & ab^* + cd^* \\ a^*b + c^*d & |b|^2 + |d|^2 \end{pmatrix}.$$

The eigenvalues of the matrices ρ_A and ρ_B are given by

$$\lambda_{1,2} = \frac{1}{2}\left(1 \pm \sqrt{1 - C^2}\right). \tag{22}$$

The concurrence C of the system with the wave function (20) is[11]

$$C = 2\,|ad - bc|. \tag{23}$$

From known the eigenvalues of the matrices ρ_A and ρ_B, one can calculate the kth moment of the entropy operator $-\log_2\rho_A$ [see expressions (6) and (7)][42]

$$\overline{S}^k = (-1)^k \left[\frac{1 + \sqrt{1 - C^2}}{2}\log_2^k \frac{1 + \sqrt{1 - C^2}}{2} + \right. \tag{24}$$
$$\left. + \frac{1 - \sqrt{1 - C^2}}{2}\log_2^k \frac{1 - \sqrt{1 - C^2}}{2}\right].$$

Note that all characteristics of entanglement of the two-qubit system are determined by the concurrence C.

Using the equalities (19) and (24), the fluctuations of quantum entanglement of the two-qubit system in a pure state[32] are

$$\Delta E = C\log_2\left[\frac{1}{C}\left(1 + \sqrt{1 - C^2}\right)\right]. \tag{25}$$

Since the quantum entanglement depends only on the concurrence and, as follows from relation (25), the same holds for the quantum entanglement fluctuations, ΔE can be measured independently from the quantum entanglement.

Changes in quantum entanglement and entanglement fluctuations upon changes in the concurrence C of the two-

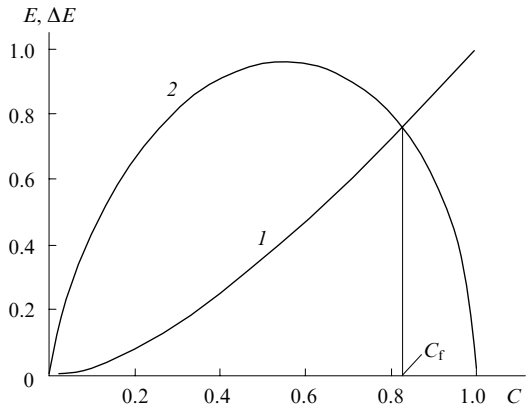


Figure 1. Quantum entanglement (*1*) and entanglement fluctuations (*2*) plotted *vs.* concurrence C in a two-qubit system in a pure state.[32] $C_f \approx 0.82724$.

qubit system are illustrated in Fig. 1. At $C = 0$ (no entanglement), the absolute fluctuations are equal to zero. At $C = 1$ (maximally entangled state), there are also no entanglement fluctuations; however, as the concurrence deviates from unity, fluctuations in the system increase and become equal to the entanglement ($\Delta E = E$) at $C = C_f$, where C_f is the root of the transcendent equation

$$\left(C_f + \sqrt{1 - C_f^2}\right) \ln \frac{1}{C_f} \left(C_f + \sqrt{1 - C_f^2}\right) = \ln \frac{2}{C_f}. \tag{26}$$

The numerical value of the root of Eqn (26) is $C_f \approx 0.82724$. At $0 < C < C_f$, the system is in the region of large-amplitude fluctuations ($\Delta E > E$). In the text below we will show that these considerations can be used for the choice of materials for quantum device applications.

An analysis [32] of quantum entanglement fluctuations for a mixed state of a two-qubit system showed that relation (25) holds in this case provided that the concurrence is determined using the Wootters relation (16).

As an example, we will consider a Heisenberg dimer with the Hamiltonian

$$H = \frac{1}{2} J \overline{\sigma}_1 \, \overline{\sigma}_2 , \tag{27}$$

where J is the exchange coupling constant and $\overline{\sigma}_i = (\sigma_{ix}, \sigma_{iy}, \sigma_{iz})$, $i = 1, 2$. The density matrix for the dimer existing in equilibrium at temperature T is [32]

$$\rho(T) = \frac{1}{Z} \exp\left(-\frac{H}{kT}\right) = \tag{28}$$

$$= \frac{1}{Z} e^{-3K} |\Psi^-\rangle\langle\Psi^-| + \frac{1}{Z} e^{K} \left(|\Psi^+\rangle\langle\Psi^+| + |\Phi^+\rangle\langle\Phi^+| + |\Phi^-\rangle\langle\Phi^-|\right),$$

where the partition function is

$$Z = 3e^{K} + e^{-3K}, \tag{29}$$

$$K = \frac{J}{2kT},$$

k is the Boltzmann constant, $|\Psi^{\pm}\rangle$, $|\Phi^{\pm}\rangle$ are the Bell states:

$$|\Psi^{\pm}\rangle = \frac{1}{\sqrt{2}} \left(|01\rangle \pm |10\rangle\right), \tag{30}$$

$$|\Phi^{\pm}\rangle = \frac{1}{\sqrt{2}} \left(|00\rangle \pm |11\rangle\right).$$

In this system, the coherence differs from zero only at $J < 0$ (antiferromagnetic case) and is given by [32]

$$C(T) = \begin{cases} -1 + \dfrac{2}{1 + 3\exp[-2|J|/(kT)]}, & T < T_e \\ 0, & T \geqslant T_e \end{cases}, \tag{31}$$

where

$$T_e = \frac{2}{\ln 3} \frac{|J|}{k}. \tag{32}$$

These expressions and relation (25) give the desired dependence of fluctuations of thermal entanglement $\Delta E(T)$ in the Heisenberg dimer.

The temperature dependences of entanglement and entanglement fluctuations in the Heisenberg dimer are shown in Fig. 2. The $E(T)$ and $\Delta E(T)$ values are equal at $T = T_f$ (see Ref. 32)

$$\frac{kT_f}{|J|} = 2 \ln \frac{3(1 + C_f)}{1 - C_f} \approx 0.57849. \tag{33}$$

From Fig. 2 it follows that at $T > T_f$, the entanglement fluctuations are larger than the entanglement values. At $T \geqslant T_e$ (in a separable state), the concurrence (31), entanglement and entanglement fluctuations are equal to zero.

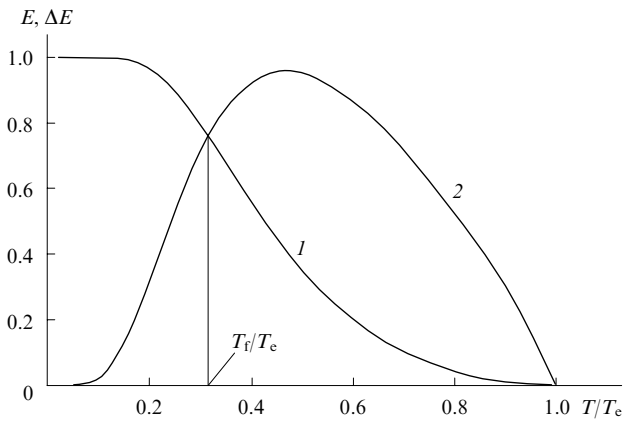


Figure 2. Quantum entanglement (*1*) and entanglement fluctuations (*2*) in the Heisenberg dimer plotted *vs.* relative temperature T/T_e.[32] $T_f/T_e = 0.31776$.

From relation (32) it follows that an increase in T_e can be attained by choosing materials in which magnetic dimers are characterized by a larger coupling constant $|J|$. The T_e value also depends on the experimental method employed. For instance, the T_e value reached in a multi-quantum NMR experiment [51] was nearly three orders of magnitude higher than the T_e obtained in an experiment using thermally equilibrium magnetization.[52]

Summing up, an analysis of the entropy of entanglement should be performed taking into account entanglement

fluctuations. The entanglement fluctuations should not be ignored when considering systems that undergo transition from an entangled to a separable state.[32] There considerations will be applied subsequently to the experimental temperature dependences of the magnetic susceptibility (witness of entanglement) in nitrosyl iron complexes.[15]

IV. Quantum correlations and quantum discord

According to the theory outlined in the preceding Sections, all quantum states can be divided into two types, *viz*., entangled and non-entangled (separable) ones. In this connection, it was assumed that quantum devices, in particular, quantum computers, can provide considerable advantages compared to their classical analogues only through quantum mechanical entanglement.[2] It was suggested[2] that acceleration of quantum calculations is possible only through the use of entanglement in quantum algorithms. For instance, algorithms implemented using liquid-phase NMR methods[53] in the systems containing a few nuclear spins (no entanglement) do not allow one to expect a significant acceleration of calculations in the experiments with a large number of nuclear spins (qubits).[36, 54] Taking into account this fact, the liquid-phase NMR methods[55, 56] used to produce a seven-qubit quantum computer on nuclear spins of crotonic acid in 2000 (Ref. 53) were recognized as inappropriate.

In fact, the situation is much more complicated than the entanglement – separability scenario, which underlies all conclusions drawn above. Generally, one should proceed from the assumption that the density matrix ρ containing complete information on a quantum mechanical system includes both classical and quantum correlations. It is quantum correlations that provide advantages of quantum devices compared to classical analogues. The main issue is to separate the two types of correlations. It should be emphasized that entanglement is not the only type of quantum correlations responsible for the advantages of quantum devices.[39] This was confirmed by recent studies. In this connection, noteworthy are investigations of 'quantum nonlocality' (no entanglement)[33–35] and a model[36–38, 57, 58] demonstrating 'quantum acceleration' on separable states. Therefore, it is of paramount importance to study all quantum correlations in the system under consideration.

Another approach to study the quantum correlations different from those characterized by entanglement was proposed independently by Henderson abd Vedral[59] and by Ollivier with Zurek.[39, 60] Their idea is based on the fact that in some cases, equivalent classical expressions lead to different quantum analogues owing to non-commutativity of operators that describe the observable states in quantum mechanics.[42] This difference can be used for quantitative description of 'quantumness' of the object under study.

Now we will consider quantitative description of quantum discord. In Shannon's theory,[48] information on a random event, as a measure of our ignorance of the event, is determined by its probability. Quantitatively, information $I(p)$ is described by Shannon's entropy $H(p)$. First, we will consider a system comprising particles A and B. Mathematically, this system is described by the probability of joint distribution of these particles p_{jk}, where j and k refer to the results of measurement of states of one-particle subsystems A and B, respectively. The probability for the subsystem A to be in the state j is given by

$$p_j^A = \sum_k p_{jk}\,, \tag{34}$$

where summation is carried out over all states k of the subsystem B. Analogously, the probability for the subsystem B to be in the state k is

$$p_k^B = \sum_j p_{jk}\,, \tag{35}$$

where summation is carried out over all states j of subsystem A. The expression for Shannon's entropy of the entire system has the form[48]

$$H(p) = -\sum_{jk} p_{jk} \log_2 p_{jk}\,. \tag{36}$$

The classical mutual information $I(p)$ is calculated as follows:

$$I(p) = H(p^A) + H(p^B) - H(p), \tag{37}$$

where $H(p^A)$ and $H(p^B)$ are the entropies of subsystems A and B, respectively. It is important that relation (37) can be represented in equivalent form

$$I(p) = H(p^A) - H(p|p^B), \tag{38}$$

where $H(p|p^B)$ is the conditional entropy

$$H(p|p^B) = H(p) - H(p^B). \tag{39}$$

The physical sense of conditional entropy $H(p|p^B)$ is as follows: it characterizes the change in the information on the system due to the change in ignorance of it owing to the fact that the state of the subsystem B is known (ignorance factor decreased). Geometric relations between the mutual entropy, conditional entropies and mutual information are illustrated in Fig. 3.

Now we will use the Bayes formulas from the probability theory[40]

$$p_{jk} = p_{j|k}\, p_k^B = p_{k|j}\, p_j^A, \tag{40}$$

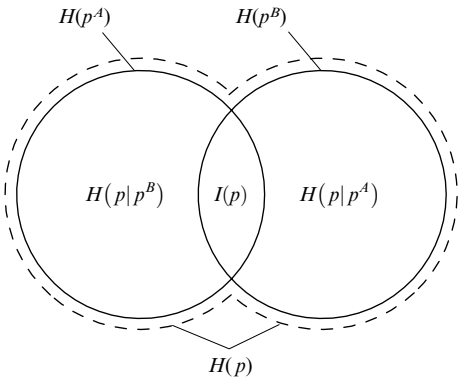


Figure 3. Relations between $H(p)$, $H(p|p^B)$, $H(p|p^A)$ and $I(p)$.

where $p_{j|k}$ is the probability for the subsystem A to be in the state j provided that subsystem B is in the state k. Taking into account relations (34)–(36) and (39), (40), one gets with ease

$$H(p|p^B) = -\sum_{j,k} p_{jk} \log_2 p_{j|k}\,. \tag{41}$$

In the quantum case, the classical probability distribution is replaced by the density operator and summation is replaced by the trace calculations. Thus, the density matrix ρ of a system comprising the subsystems A and B plays the role of the joint probability p and the probabilities p^A and p^B of the states of the subsystems are replaced by the reduced density matrices

$$\rho^A = \mathrm{Tr}_B\{\rho\},$$

$$\rho^B = \mathrm{Tr}_A\{\rho\}.$$

The functional of the Shannon entropy is replaced by the von Neumann quantum entropy [61]

$$S(\rho) = -\mathrm{Tr}\{\rho \log_2 \rho\}. \tag{42}$$

This correspondence enables generalization of the classical mutual information to the quantum case. It naturally begins with the introduction of quantum mutual information:

$$I(\rho) = S(\rho^A) + S(\rho^B) - S(\rho) = S(\rho^A) - S(\rho|\rho^B), \tag{43}$$

where the quantum conditional entropy $S(\rho|\rho^B)$ [generalization of the classical conditional entropy (39) to the quantum case] is given by:

$$S(\rho|\rho^B) = S(\rho) - S(\rho^B). \tag{44}$$

The quantum mutual information has a fundamental physical sense and is used as a measure of total correlations.[62–69]

To reveal quantum correlations in the system, Ollivier and Zurek [39] considered another procedure for extension of the classical mutual information to the quantum case. Their idea was to determine the quantum conditional entropy using measurements for one subsystem of the system under study (for instance, subsystem B). Here, the term 'measurement' is always treated in the von Neumann sense as containing a set of one-dimensional projectors whose sum is equal to unity.[70] Let $\{B_k\}$ be a complete set of such measurements carried out only for the subsystem B. Then, the quantum state of the system after kth measurement is described by the following density matrix ρ_k:

$$\rho_k = \frac{1}{p_k}(I \otimes B_k)\rho(I \otimes B_k), \tag{45}$$

where I is the unity operator in the Gilbert space of the subsystem A and the probability p_k of the kth state is

$$p_k = \mathrm{Tr}(I \otimes B_k)\rho(I \otimes B_k). \tag{46}$$

From Eqn (46) one gets

$$\sum_k p_k = 1.$$

The density matrix ρ_k (conditional density matrix) can be treated as formal quantum generalization of the classical conditional probability distribution.[62,71] Using this density matrix, one gets a new expression for the quantum conditional entropy, which provides an alternative to relation (44):

$$S(\rho|\{B_k\}) = \sum_k p_k S(\rho_k), \tag{47}$$

where $\{B_k\}$ is the complete set of measurements for the subsystem B. For classical systems, expressions (44) and (47) are equivalent, but this is not the case for quantum systems. The reason is that, generally, it is impossible to perform quantum measurements for subsystem B without influencing subsystem A. An analogue of quantum mutual information obtained using the complete set of measurements $\{B_k\}$ for subsystem B can be defined as follows:

$$I(\rho|\{B_k\}) = S(\rho^A) - S(\rho|\{B_k\}). \tag{48}$$

Emphasize that the mutual information $I(\rho|\{B_k\})$ is determined after a complete set of measurements was done for the subsystem B, *i.e.*, when it becomes classical. Therefore, it is appropriate to relate $I(\rho|\{B_k\})$ to the classical correlations. As a measure of classical correlations, Ollivier and Zurek [39] introduced the quantity

$$C(\rho) = \sup_{\{B_k\}} I(\rho|\{B_k\}), \tag{49}$$

where the maximum $C(\rho)$value is taken over all all possible complete sets of measurements for subsystem B.

Thus, we have two quantum analogues of mutual information, namely, the quantum mutual information [see relation (43)] which is a measure of total correlations[2] and the mutual information [see expression (49)] which is a measure of classical correlations. The difference between these mutual correlations

$$Q(\rho) = I(\rho) - C(\rho), \tag{50}$$

is called the quantum discord;[39] it was proposed it as a measure of quantum correlations.

The quantum discord is a non-negative quantity.[39] Owing to a complicated optimization procedure included in the definition of quantum discord it cannot always be calculated in explicit form. However, methods for the determination of the quantum discords for two-qubit systems whose density matrices represent X-matrices have been developed.[72,73] This allows one to compare the total, quantum and classical correlations.

A comparison of the quantum discord with quantum entanglement was reported for some two-qubit systems.[72,73] The quantum discord can be either larger or smaller than the entanglement.[72,73] This means that the measures of quantum correlations (quantum entanglement and quantum discord) differ not only quantitatively, but also qualitatively.

The definition of quantum discord as a measure of quantum correlations has certain drawbacks. For instance, the quantum discord depends on which subsystem was used

in measurements.[39] The asymmetry of the quantum discord was studied using a simple two-spin model.[74] It was shown that asymmetry can be as high as 25% of the quantum discord. Because of this, a symmetric version of determination of bipartite quantum discord based on two-side projective measurements was proposed.[75]

The introduction of quantum discord led to fundamental changes in the strategy for the search for materials for quantum device applications, namely, the efficiency of operation of quantum devices is determined by quantum correlations (this is in line with the general methodology of research on quantum systems) rather than the 'entangled–separable state' scenario.

V. Experimental studies of quantum entanglement

After successful experiments with particles (protons, photons and, more recently, mesons) to validate the Bell inequalities carried out in the 1970s–1980s to prove the existence of entanglement between particles, researchers undertook attempts to disclose quantum correlations in condensed media.

In 1997, it was reported[76] that an anomaly in neutron scattering from H_2O-D_2O mixture represents 'direct evidence of short-lived entanglement of adjacent protons (deuterons) in condensed matter at room temperature'. However, this conclusion cast doubt upon.[77]

More recenly, an experimental study of salt $LiH_xY_{1-x}F_4$ at ultralow temperatures (<1 K) revealed deviations of the temperature dependence of the magnetic susceptibility from the Curie law ($\chi \sim 1/T$), which were attributed to quantum entanglement.[14]

A new stage in proving the existence of quantum entanglement in various materials began with the study by Brukner, Vedral and Zeilinger,[16] who developed a solid theoretical foundation for identification of quantum entanglement. They studied crystalline copper nitrate hydrate $Cu(NO_3)_2 \cdot 2.5\,H(D)_2O$. The magnetic structure of this compound represents antiferromagnetic one-dimensional chains of Heisenberg spins ($s = 1/2$) with alternant interactions of the nearest neighbours. Using the dimer model, the entanglement temperature estimated from the magnetic susceptibility and neutron-scattering data is $T_e \approx 5$ K. Owing to neat arrangement of arguments the study[16] gave an impetus to new investigations.

The $(NHEt_3)[V^{IV}_8V^{V}_4As_8O_{40}(H_2O)] \cdot H_2O$ molecular antiferromagnet was studied.[17] Using experimental values of the magnetic susceptibility, it was found that for this compound $T_e \approx 25.4$ K. In this case, the intersection of the magnetic susceptibility curve with the Curie hyperbola was used as the witness of entanglement.

An analysis of experimental data on the magnetic susceptibility of nanotubular crystal $Na_2V_3O_7$ built of nanotubes consisting of weakly coupled nine-site rings.[18] Using the same indicator of entanglement, it was concluded that thermal entanglement in the material occurs at temperatures below 365 K.

The magnetic susceptibility measurements for two magnetic systems, pyroborate $MgMnB_2O_5$ (spin $s = 5/2$) and varvicite $MgTiOBO_3$ (spin $s = 1/2$) were carried out.[19] Using the temperature dependences of the magnetic susceptibility as witness of entanglement, it was found that $T_e \approx 18$ K for $MgMnB_2O_5$ and 100 K for $MgTiOBO_3$.

A neutron scattering study of crystals of organometallic magnet $Me_2NH_2 \cdot CuCl_3$ revealed quantum correlations (entanglement) in them.[20] However, no quantitative estimations values of entanglement or at least T_e were presented.

A T_e value of ≈ 240 K was reported[21] for crystalline oxide $Na_2Cu_5Si_4O_{14}$ containing pentanuclear Heisenberg clusters of Cu^{2+} ions based on the available experimental data on the magnetic susceptibility of this compound.

Nitrosyl iron complexes $[Fe_2(C_3H_3N_2S)_2(NO)_4]$ (**1**) and $[Fe_2(SC_3H_5N_2)_2(NO)_4]$ (**2**) were studied.[15] Using the magnetic susceptibility data, quantum mechanical entanglement of the spin degrees of freedom was revealed for both complexes. The T_e values are 80–90 K for **1** and 110–120 K for **2**. In addition, the temperature dependence of quantum entanglement for complex **2** was determined using the relation between the entanglement and magnetic susceptibility. A detailed analysis of quantum entanglement, its fluctuations and quantum discord in the nitrosyl iron complexes will be presented in the text below.

The existence of entanglement was also established for the *syn–syn* conformation of copper carboxylate $[Cu_2(O_2CH)_4][Cu(O_2CH)_2(Pic)_2]$ (Pic is 2-methylpyridine).[22] The magnetic structure of this compound comprises copper dimers and monomers. Using a known correlation between concurrence and the magnetic susceptibility of a Heisenberg dimer, a record-high T_e of 630 K was obtained for copper carboxylate. The temperature dependence of concurrence was also reported.[22]

The entanglement temperatures of a series of isostructural oxides $KNaMSi_4O_{10}$ (M = Mn, Fe, Cu) were determined.[23] Estimation was also made based on the witness of entanglement constructed using the magnetic susceptibility data. These oxides have dimeric magnetic structures, but the spin values and the character of interactions in them are different. Namely, copper oxide contains antiferromagnetic dimers with $s = 1/2$, iron oxide contains ferromagnetic dimers with $s = 2$, and manganese oxide contains antiferromagnetic dimers with $s = 5/2$. The witness of entanglement revealed $T_e \approx 2.43(7)$ and 8.91(9) K for the copper and manganese oxide, respectively. Owing to the ferromagnetic character of interactions in the iron oxide crystal, no entanglement occurs in that material.

An entanglement study of a complex $Cr_7Ni-Cu^{2+}-Cr_7Ni$ comprising two Cr_7Ni rings linked by a copper ion was carried out.[24] At low temperatures, each Cr_7Ni ring can be reduced to an effective system with the spin $s = 1/2$ (qubit). It is important that such a qubit is characterized by a long decoherence time and can therefore be considered as a promising material for devices for quantum information processing. This complex can quantitatively be described by a Heisenberg trimer. The behaviour of pair entanglements between the central ion (Cu^{2+}) and one ring and between ring-qubits Cr_7Ni was calculated for this system.

Recently, attempts to establish the role of quantum correlations in biological systems at physiological temperatures have been undertaken (see Ref. 25 and references cited therein). A real candidate for being used in such systems is a Fenna–Matthews–Olson protein light-harvesting complex. However, investigations in this field are mainly theoretical.

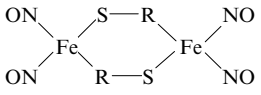
The entanglement temperatures for the compounds mentioned above are shown in Table 1.

Table 1. The entanglement temperatures of various compounds.

Compound	Characteristic of system	T_e/K	Ref.
$LiHo_xY_{1-x}F_4$	diluted ferromagnet	< 1	14
$Cu(NO_3)_2 \cdot 2.5\,H(D)_2O$	alternant antiferromagnetic 1D chain	5	16
$(NHEt_3)[V_8^{IV}V_4^{V}As_3O_{40}(H_2O)]H_2O$	molecular antiferromagnet	25.4	17
$Na_2V_3O_7$	nanotubes	365	18
$MgMnB_2O_5$	$s = 5/2$	18	19
$MgTiOBO_3$	$s = 1/2$	100	19
$Me_2NH_2CuCl_3$	organic magnet, $s = 1/2$, alternant 1D chains		20
$Na_2Cu_5Si_4O_{14}$	pentanuclear Heisenberg clusters	240	21
$Fe_2(C_3H_3N_2S)_2(NO)_4$	Heisenberg dimer	80 – 90	15
$Fe_2(SC_3H_5N_2)_2(NO)_4$	the same	110 – 120	15
$[Cu_2(O_2CH)_4][Cu(O_2CH)_2(Pic)_2]$	dimers + monomers	630	22
$KNaMnSi_4O_{10}$	$s = 5/2$	8.91(9)	23
$KNaFeSi_4O_{10}$	$s = 2$	0	23
$KNaCuSi_4O_{10}$	$s = 1/2$	2.43(7)	23
$Cr_7Ni - Cu^{2+} - Cr_7Ni$	two Cr_7Ni rings linked by Cu^{2+} ion (three-qubit molecular system)		24
The Fenna – Matthews – Olson protein light-harvesting complex	biomolecular complex	–	25

VI. Quantum entanglement, fluctuations of quantum entanglement and quantum discord in binuclear nitrosyl iron complexes

Nitrosyl iron complexes **1** and **2** contain nitrogen monoxide, a signal molecule in some metabolic and physiological processes. In tissues of living organisms, these complexes were found in the 1960s.[78] They exhibit a characteristic EPR signal with a g-factor of 2.03 and were thus called the 'complexes 2.03'. Unfortunately, natural binuclear nitrosyl iron complexes are very unstable. However, their properties can be modelled using artificial analogues.[79] The structure of the binuclear complexes in question can be described as follows:



(51)

R is imidazol-2-yl (**1**) or imidazolin-2-yl (**2**).

The substituents R represent five-membered heterocycles containing three carbon atoms and two nitrogen atoms separated by a carbon atom.

Studies of nitrosyl iron complexes by X-ray diffraction analysis, Mössbauer spectroscopy and SQUID magnetometry showed the following.[80 – 82] The magnetoactive centres in the complexes are formed by iron ions; each iron ion is bound to two nitrosyl groups. Each magnetoactive centre has a spin of 1/2. The molecules have a centrosymmetric structure in which both iron atoms are linked by S – C═N bridges, where C – N is a fragment of the heterocycle R. In the paramagnetic dimer of the nitrosyl complex, spins are coupled through superexchange antiferromagnetic interaction. The Fe – Fe distance is 0.4102 (**1**) and 0.4030 (**2**) nm.

Crystals of the complexes in question have a layered structure. Within the layer, the aligned molecules form intermolecular contacts between a sulfur atom and H – N fragment of the substituent R. Owing to long lengths of these superexchange bonds, the dimer – dimer interactions are weak. Different layers are linked through electrostatic forces only, which also favours separation of dimers from one another.

Experiments showed that, magnetically, the binuclear complexes **1** and **2** represent almost free antiferromagnetic Heisenberg dimers.[15]

The magnetic susceptibility $\chi(T)$ per mole of Heisenberg dimers is described by the Bleaney – Bowers equation [83]

$$\chi(T) = \frac{2N_A g^2 \mu_B^2}{kT[3 + \exp(-2J/kT)]}, \tag{52}$$

where N_A is the Avogadro constant; μ_B is the Bohr magneton; g is the corresponding component of the g-factor if measurements are carried out for a single crystal, or

$$g^2 = \frac{g_x^2 + g_y^2 + g_z^2}{3}, \tag{53}$$

if polycrystalline (powder) samples are used. Using Eqn (52), from relation (31) one gets the following expression for the concurrence of antiferromagnetic Heisenberg dimer (see relationship (27)) at $T < T_e$:

$$C(T) = 1 - \frac{3}{2}\frac{\chi(T)}{\chi_C(T)}, \tag{54}$$

where

$$\chi_C(T) = \frac{N_A g^2 \mu_B^2}{2kT} \tag{55}$$

is the Curie magnetic susceptibility for two spins $s = 1/2$ [Eqn (52) at high temperatures where one can say that $J = 0$]. Using these relations, expressions (13) and (14) and the experimentally measured magnetic susceptibility values, one can determine the quantum entanglement of the system of Heisenberg dimers.

According to expression (54), quantum entanglement in dimer (27) exists if

$$\chi(T) < \frac{2}{3}\chi_C(T). \tag{56}$$

Inequality (56) is consistent with a more general criterion for non-separability, *viz*., nonzero quantum entanglement in the system of N particles with the spins s:[84]

$$\chi_p(T) < \frac{Ng^2\mu_B^2 s}{3kT}, \tag{57}$$

where χ_p is the magnetic susceptibility averaged over spatial directions (susceptibility of the polycrystalline material).

For comparison with experimental data, it is convenient to write relation (57) in the form

$$\chi_p(T) < \frac{1}{1+s}\,\chi_C(T), \tag{58}$$

where

$$\chi_C(T) = \frac{nN_A g^2\mu_B^2 s(s+1)}{3kT} \tag{59}$$

is the Curie law for a mole of n-nuclear clusters comprised of the spins s (in dimer, $n = 2$).

The right parts of the inequalities (56) and (58) represent the modified [owing to the coefficients 2/3 and $1/(1+s)$] Curie laws. Having determined the coordinates of the intersection points of these curves with the temperature dependences of the magnetic susceptibility, one can determine the temperature ranges in which the dimer is in the entangled and separable states.

The behaviour of the initial magnetic susceptibility of complex **1** is shown in Fig. 4.[80] The sample studied contained a small amount of impurities ($\sim$2.3%), which contribute to the increase in the magnetic susceptibility near $T = 0$ K. At $T > T^{\chi}_{max}$ (T^{χ}_{max} is the temperature corresponding to the maximum magnetic susceptibility value), the contribution of impurities is relatively small and we will neglect it in the estimation of T_e (**1**).

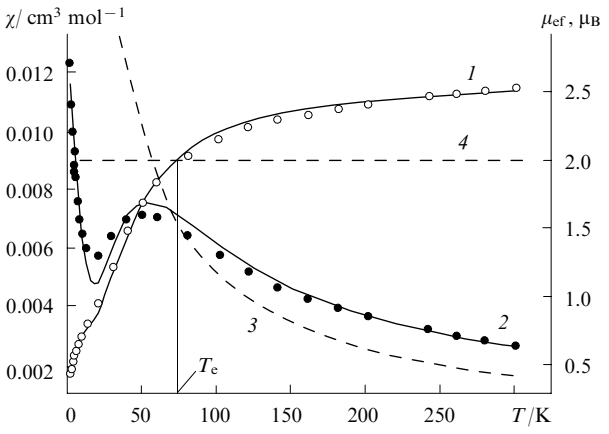


Figure 4. Temperature dependences of the effective moment (*1*) and magnetic susceptibility (*2*) of complex **1**.
The Figure was taken from Ref. 80 and augmented with the dashed lines *3* and *4* (see text). $T_e \approx 80$ K.

The dashed line *3* in Fig. 4 represents the modified Curie hyperbola [right part of inequality (56)] taking into account the fact that for the complex under study one has $g = 2$.[80] This hyperbola intersects the experimental dependence (filled circles) at $T_e \approx 80$ K. Estimation from the maximum of the magnetic susceptibility[15] gives $T_e \approx 90$ K. This suggests that quantum entanglement in complex **1** emerges at $T < 80-90$ K.

In magnetochemistry, it is commonly accepted to represent the same experimental data in the form of $\chi(T)$ curves and as the temperature dependence of the effective magnetic moment

$$\mu_{ef}(T) = \frac{1}{\mu_B}\left(\frac{3k}{N_A}T\chi\right)^{1/2}. \tag{60}$$

Then, criterion (58) can be written as follows:

$$\mu_{ef}(T) < g\sqrt{ns}. \tag{61}$$

To reveal the presence or absence of entanglement, one should draw a horizontal straight line at $g\sqrt{ns}$, look whether it intersects the $\mu_{ef}(T)$ curve and determine the coordinates of the intersection point (if exists). In Fig. 4, the horizontal dashed line *4* is drawn at a level of $g\sqrt{ns} = 2$ ($g = 2$, $n = 2$ and $s = 1/2$). The abscissa of the intersection point of this line with the plot of the temperature dependence of μ_{ef} is consistent with the temperature T_e found above.

An analysis of entanglement in complex **2** is illustrated in Fig. 5. To carry out a more thorough study of entanglement, we substracted the impurity contribution from $\chi(T)$ and thus obtained the temperature dependence of the magnetic susceptibility of complex **2** *per se*. Curve *3* in Fig. 5 is the Bleaney–Bowers plot [see Eqn (52)] with the parameters taken from Ref. 81, namely, $J/k = -68$ K and $g = 2$. The modified Curie curve intersects the temperature dependence *2* at $T_e \approx 110$ K. Estimation of this temperature from the maximum of the $\chi(T)$ curve gives $T_e \approx 121$ K.[15] Thus, one has T_e(**2**) $\approx 110-120$ K.

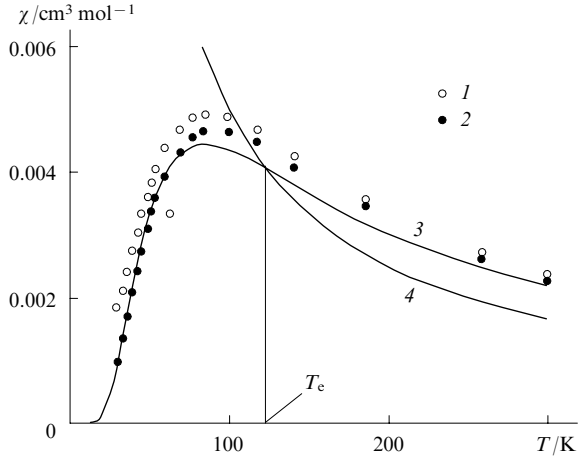


Figure 5. Temperature dependences of the magnetic susceptibility of complex **2** with inclusion of the impurity contribution (*1*) and after subtraction of this contribution (*2*).
The Bleaney–Bowers theoretical dependence with $J/k = -68$ K and $g = 2$ (*3*) and the $(2/3)\chi_C(T)$ dependence with $g = 2$ (*4*). Experimental data denoted by the open and solid circles were taken from Ref. 81.

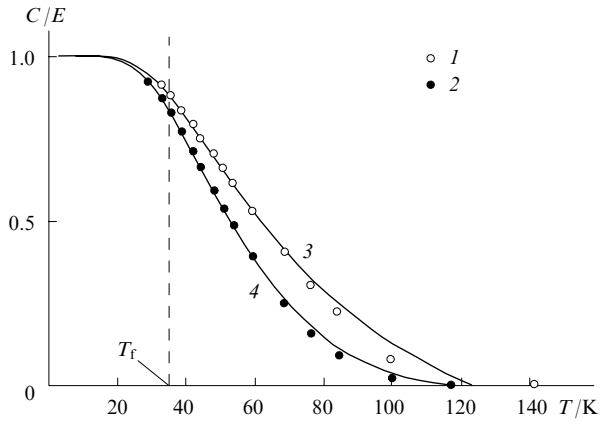


Figure 6. Temperature dependences of concurrence (*1*) and entanglement (*2*) for complex **2**.[15]
Solid curves *3* and *4* represent the theoretical dependences of C and E, respectively. The dashed line corresponds to $T_f = 38$ K; at higher temperatures, fluctuations of quantum entanglement are larger than the entanglement.

Figure 6 represents the temperature dependences of the coherence and entanglement recalculated using expressions (54), (55), (13) and (14) from the experimental data [81] for the magnetic susceptibility of complex **2**. Solid curves denote the theoretical dependences $C(T)$ and $E(T)$ obtained using the Bleaney–Bowers equation (52) and the given above J/k and g values for **2**. The data shown in Fig. 6 suggest that quantum entanglement in complex **2** is as high as 0.90–0.95 at 25 K.

Now we will dwell on the physical nature of entanglement in nitrosyl iron complexes.[85] The Heisenberg dimer [see expression (27)] has two energy levels, namely, $3J/2$ and $-J/2$, corresponding to the singlet and triplet states, respectively. The singlet state is the ground state of antiferromagnetic dimer at $T = 0$ K. The entanglement of the antiferromagnetic dimer at $T = 0$ K equals to unity (maximally entangled state). As temperature increases, the energy level of the triplet state begins to populate. This is accompanied by a decrease in entanglement. Entanglement vanishes when the statistical weight of the singlet state in the density matrix decreases to 1/2 (see Ref. 15). This occurs at $T_e \sim \Delta/k$, where $\Delta = -J/2 - 3J/2 = 2|J|$ is the energy gap in the spectrum. At $T > t_e$, the system exists in the separable state.

In Section III, we considered fluctuations of quantum entanglement. The temperature dependence of concurrence [Eqn (31)] can be used to determine the temperature dependences of entanglement and its fluctuations (see Fig. 2). It was found [32] that at $T_f = 0.31776\, t_e$, fluctuations of quantum entanglement become equal to the entanglement.

Evaluation of fluctuations of a measure of quantum correlations (in particular, quantum entanglement) is of crucial importance when elucidating the applicability of materials to the design of quantum devices. The case in point is that the potential of quantum devices having no classical analogues is due to quantum correlations governed by the quantum discord (this is impossible for classical physics).[2] In some cases, the appropriateness of a material for quantum device applications can be assessed from the quantum entanglement standpoint. For instance, a register of a quantum computer based on chains of nuclear spins should include entangled states of an arbitrary pair of spins (qubits). In this case, one can attain an exponential increase in perfomance when solving some complicated tasks that cannot be solved using classical computers.[2] In estimation of pair entanglement one should take into account its fluctuations. The magnitude of entanglement suitable for the use of nitrosyl iron complexes as materials for quantum device applications is attained at temperatures below $T_f = 38$ K (see Fig. 6). The vertical straight line separates the temperature ranges with small- ($T < T_f$) and large-amplitude ($T > T_f$) fluctuations of entanglement. At $T_f \approx 38$ K, fluctuations of entanglement in complex **2** become of the order of the entanglement.

Consider the behaviour of the quantum discord. For the Hamiltonian (27), the density matrix (28) can be written in the form [86]

$$\rho(T) = \frac{1}{4}(1 + G\,\bar{\sigma}_1\,\bar{\sigma}_2) = \tag{62}$$

$$= \frac{1}{4}\begin{pmatrix} 1+G & 0 & 0 & 0 \\ 0 & 1-G & 2G & 0 \\ 0 & 2G & 1-G & 0 \\ 0 & 0 & 0 & 1+G \end{pmatrix},$$

where

$$G(T) = -1 + \frac{4}{3 + \exp(-2J/kT)}. \tag{63}$$

The quantum discord (Q) in the system described by the density matrix (62) can be calculated exactly:[72]

$$Q = I - C. \tag{64}$$

In this case, the mutual information is

$$I = \frac{1}{4}\left[(1-3G)\log_2(1-3G) + 3(1+G)\log_2(1+G)\right], \tag{65}$$

and the classical part of total correlations is

$$C = \frac{1}{2}\left[(1+|G|)\log_2(1+|G|) + (1-|G|)\log_2(1-|G|)\right]. \tag{66}$$

Using Eqn. (52), one gets

$$G(T) = \frac{2kT\chi(T)}{N_A g^2 \mu_B^2} - 1. \tag{67}$$

Expressions (63)–(67) allow the quantum discord to be calculated from the magnetic susceptibility data for a compound with the magnetic structure built of Heisenberg dimers. The temperature dependence of the quantum discord is illustrated by Fig. 7. At 0 K, the quantum discord, as well as quantum entanglement, is maximum, *viz.*, $Q = 1$. As the temperature increases, the quantum discord initially changes only slightly (nearly horizontal portion of the curve $Q(T)$ at $T < 20$ K). Further increase in the temperature causes the quantum discord to decrease and at high temperatures, one gets $Q \sim 1/T^2$. It should be noted that, in contrast to entanglement, the quantum discord differs from zero at all temperatures.

Dimers are model systems for studies of quantum discord, quantum entanglement and its fluctuations. Evalu-

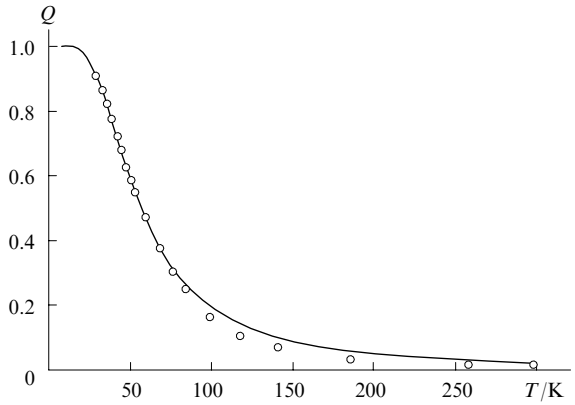


Figure 7. Temperature dependence of quantum discord for complex **2**.
Open circles denote experimental data of magnetic susceptibility measurements and the solid curve is the theoretical dependence.

ation of quantum discord and entanglement of multi-spin (multi-qubit) systems is important for the design of materials for quantum devices. Theoretically, the problem is to develop a theory of quantum discord and entanglement for multi-qubit systems. Using this theory and theory of fluctuations of the quantum discord and entanglement, one can elaborate a reliable test procedure to assess the appropriateness of materials for quantum device applications, in particular, for production of quantum computers.

VII. The quantum state transfer

The problem of ideal transfer of a quantum state or high-probability quantum state transfer [87, 88] is immediately related to the problem of quantum teleportation.[89–93] In essence, transfer of a quantum state along long chains comprising 1000 and 10 000 atomic nuclei with nonzero spins was observed in the numerical experiments with the so-called quantum echo or Loschmidt echo,[94] although the transfer probability in these experiments is low.[95] The problem of ideal transfer of a quantum state along a uniform open-end Heisenberg chain in the nearest neighbour interaction approximation was for the first time considered by Bose,[87] who reported that efficient quantum state transfer is possible only along short uniform chains. At present, main efforts of researchers are aimed to implement the quantum state transfer between chain ends, which is typical for quantum communication. It was shown that symmetric nonuniform chains are most promising to this end.[96–101] A number of methods for long-range transfer of an arbitrary quantum state have been proposed. Among them, the best known methods are as follows.

1. An ideal transfer is possible along a symmetric nonuniform chain in the nearest neighbour interaction approximation. In this case, the coupling constants increase from the chain ends towards the centre obeying some rule. In particular, a chain with the XY Hamiltonian was considered [this Hamiltonian describes isotropic spin–spin interaction in the (X, Y) plane normal to strong external magnetic field]. The coupling constants in the chain vary as follows:

$$J_n = \frac{\lambda}{2}\sqrt{n(N-n)},$$

where λ is an arbitrary constant (reciprocal time), n is the number of the node in the chain and N is the number of spins in the chain. The scatter of the coupling constant values may be large. In another case,[99] the coupling constants vary in a more complex manner, but their values are less scatterred. For instance, the scatter was 3% for a chain with $N = 31$. In both cases, ideal transfer of an arbitrary state takes the time equal to π/λ irrespective of the chain length.

2. An alternant chain in which spins are described by the XY Hamiltonian,[98, 88] in the nearest neighbour interaction approximation executes high-probability quantum state transfer. In this case, the transfer time increases with increasing the chain length.

3. High-probability quantum state transfer is possible along a chain in which all inner nuclei are at the same distance from one another and the nuclei at the chain ends are far from their neighbours (uniform chain with distant ends). Such a chain with the XXZ Hamiltonian (it describes the secular part of dipole–dipole interaction) was studied taking into account dipole–dipole interactions of all nodes.[100] A drawback of this chain consists in rapid increase in the transfer time with increasing the chain length.

4. High-probability quantum state transfer along a uniform chain with oscillating magnetic field at chain ends [102] or with inhomogeneous magnetic field along the chain.[103] The transfer time increases with the chain length.

5. Ideal quantum state transfer using 'information flux' [104, 105] in which information transferred to the jth node of the chain of atomic nuclei with the spin $s = 1/2$ is determined by the projection of the magnetization of this node.

A drawback of the first three methods consists in the need of fine 'tuning' of coupling constants for efficient transfer of states, but this is practically an egg-dance. The fourth method is free from this drawback because it considers a uniform chain placed in inhomogeneous magnetic field instead of a nonuniform chain (with different distances between neighbouring nuclei).

These quantum state transfer methods based on the use of nonniform chain and/or inhomogeneous magnetic field can complement one another depending on the problem specified or available materials. The advantage of the method based on the use of inhomogeneous magnetic field consists in the possibility of quantum state transfer to the inner nodes of the chain and in realization of entanglement between arbitrarily chosen nodes.[103] This type of problems arises in the quantum devices that distribute information between different users. In the last-mentioned method, information on the state being transferred is extracted using local operations at the chain input and output. The advantage of this method consists in that it is 'insensitive' to both the Hamiltonian and the initial state of the chain.

Relations between the quantum state transfer between nodes of a chain and the entanglement of these nodes was studied by different authors.[87, 101, 106] It was shown that the time instants corresponding to the maximum transfer probability differ from those of the maximum entanglement, except quantum state transfer along a uniform chain with distant nuclei at ends [100] (in this case, the time instants coincide).

VIII. Conclusion

We have considered important theoretical concepts underlying new quantum technologies. The development of quantum technologies, as well as fabrication of quantum devices and devices for new metrology methods is intimately related to quantum correlations that are responsible for the advantages of quantum devices over their classical analogues. The emphasis was placed on the conditions for emergence of quantum correlations and their evolution to be sufficient for the operation of quantum devices. The main condition for the existence of most quantum correlations in a system consists in the presence of quantum entanglement and small-amplitude fluctuations of the entanglement at relatively high temperatures. However, quantum entanglement does not characterize all possible (and useful) quantum correlations. The role of the total measure of all quantum correlations in the system is played by the quantum discord.[39] For pure quantum states which are often used, the quantum entanglement and quantum discord values are equal.[107] However, the quantum discord can differ from zero in mixed separable states with no entanglement. This indicates that quantum correlations, which can be used in quantum devices, are also possible in zero-entanglement systems.[33–38]

The problem of quantum decoherence[2] significantly complicates the situation. Methods for significant reduction of the effect of decoherence and to extend the lifetimes of the quantum coherencies and entangled states are developed.[108–110]

Recently, successful studies on methods of quantum cryptography,[111, 112] quantum teleportation,[89–93] as well as research on the design of quantum communication systems[113] have been carried out. Considerable advances were made in elaboration of quantum registers[26–31] and transfer of quantum information.[87, 96, 97, 99, 100, 101, 103, 106, 114] The simplest registers can be realized using materials containing long chains of atomic nuclei with nonzero spins, for instance, in chains of hydroxyl protons in hydroxyapatite $Ca_5(OH)(PO_4)_3$ (see Refs 115 and 116) or in chains of fluorine atoms in fluoroapatite $Ca_5F(PO_4)_3$.[117] Here, it is important to use both types of chains, *viz*., nonuniform (with different distances between the nearest nuclei)[96–101, 106, 114] and uniform chains placed in a specially created inhomogeneous magnetic field.[103] The idea of nonideal quantum state transfer[87, 88] (high-probability quantum state transfer with $p > 90\%$) appeared to be useful. This made it possible to implement transfer of quantum states not only between both chain ends, but also between all nodes of the chain.[118–120]

Recent advances in quantum theory of information, quantum cryptography, quantum teleportation and in experimental research on quantum entanglement gives hope for rapid progress in quantum technologies, long-awaited technologies of the future.

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