MAGNETIC QUANTUM PHASE TRANSITIONS IN DIMERIZED ANTIFERROMAGNETS

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A magnetic quantum phase transition (QPT) induced by a magnetic field in the system of exchange-coupled spin $S = 1/2$ dimers, forming a singlet in the ground state, has been considered theoretically. It has been shown that the transition of the system into a quantum antiferromagnetic (AFM) phase under the action of a magnetic field has a critical behavior. A spontaneously appeared spin polarization of the dimer is the order parameter of this phase transition. It has been shown that the absolute values of average spins of the sublattices in the quantum AFM phase depends on the magnetic field, whereas the magnetic susceptibility remains constant. It has also been demonstrated that, in the framework of the isotropic spin-spin interaction model, a transition between the quantum AFM and Néel phases is possible.

1. Introduction

An interest in the research of QPTs and, in particular, magnetic QPTs occurring at the temperature $T = 0$ has considerably grown recently [1–6]. It is related, first of all, to their difference from ordinary phase transitions; the latter occur at finite temperatures as a consequence of the ordering (or the spontaneous lowering in a symmetry) in systems with classical fluctuations. Magnetic QPT are observed, for instance, in van Vleck antiferromagnets with easy-plane single-ion anisotropy and the spin $S = 1$ of magnetic ions [7–9]. In such magnets, owing to the strong single-ion anisotropy, the ground state is a singlet state—to be exact, a state with zero spin projection. Under the action of an external field (or the temperature), the ground nonmagnetic state of magnetic ions in them gets polarized, and the whole system undergoes a spontaneous phase transition into a magnetically ordered state. The authors of works [10,11] showed that the thermodynamic approach with the introduction of the spin polarization of the ion's ground state as an order parameter is applicable to describing the QPTs in easy-plane magnets with high single-ion anisotropy.

Magnetic QPTs are phase transitions of the order–order type, because only the ground state changes in them, without any reconstruction of the ionic spectrum, the latter being characteristic of phase transitions of the order–disorder type. However, in some cases–in particular, in the dimerized singlet magnets with the common formula $XCuCl_3$, where X=(K, Cs, Tl, NH$_4$), and the spin $S = 1/2$ at every site (see review [6] and the literature therein)—the description of a magnetic QPT is complicated: difficulties are faced with, when describing both the ground singlet state and the process of its polarization.

For instance, in TlCuCl$_3$, one of the most popular compounds of this type, the structure includes pairs (dimers) of AFM-coupled spins of copper ions with $S = 1/2$ [12]. As a result, this magnet is not in the quasi-classical Néel state at $T = 0$, but in a quantum-disordered one, where the average value of spin projection is equal to zero for every ion. This state differs from a truly paramagnetic one, where the zero value of spin projections is governed by the temperature, in that this state is formed at $T = 0$. A magnet in such a state is referred to as a *quantum paramagnet.*

An external magnetic field that exceeds 23 T transforms the TlCuCl$_3$ system into a magnetically ordered state. In such a field, a spontaneous spin polarization of Cu^{2+} ions arises, and a field-induced AFM structure with canted spin vectors in its sublattices is formed. In works [5, 6], a theoretical description of such a phase transition in TlCuCl$_3$ and related compounds was made with invoking the ideas of magnon Bose–Einstein condensation (BEC). In our opinion, this phenomenon, though could be associated, by analogy, with transitions of a similar type, has no relation to the effect under consideration.

Really, in works [12–14] (for more details, see work [6]), a magnetically induced phase transition between

a singlet and a quantum, in essence, AFM state was explained by introducing a concept of BEC of a soft magnon mode. The matter is that, actually, the occurrence of transverse, with respect to the direction of an external field, projections of magnetization in finite magnetic fields can be formally described in terms of the condensation of magnetic excitations, when the energy of excitations goes to zero in the transition region. However, there is no real quasiparticle condensation in those systems, because only the reconstruction of the ground state and, as a result, virtual rather than real magnons are dealt with [15]. Otherwise, any self-consistent reconstruction–in particular, the creation of polarons, fluctuons, *etc.*–should be classed as BEC.

The consideration will be carried out using, as a guide, experimental data for mainly $TlCuCl_3$ [5,6], where the AFM exchange interaction between copper ion spins is realized. The pairs composed of the nearest Cu^{2+} ions (more precisely, Cu_2Cl_6 dimers) are structurally distinguished, because the exchange interaction between them is the strongest. The exchange between the spins of next dimers is also antiferromagnetic, but it turns out weaker. Such a hierarchy of exchange interactions brings about the situation, where the ground state of Cu^{2+} ions in every dimer is singlet, while the total spin of a dimer $S_{dim} = 0$.

Hence, the main purpose of this work is to develop the theory of spontaneous spin polarization of dimers under the action of a magnetic field in the case where every of their total spins equals zero in the initial state.

2. Model

The Hamiltonian of the system of dimers with AFM exchange between spins in every dimer and between spins of neighbor dimers (see Fig. 1) looks like [4–6]

$$\hat{H} = J_0 \sum_{\mathbf{n}_1, \mathbf{n}_2} \mathbf{S}_{\mathbf{n}_1} \mathbf{S}_{\mathbf{n}_2} + \frac{J}{2} \sum_{\mathbf{n}_\alpha, \mathbf{m}_\alpha} \mathbf{S}_{\mathbf{n}_\alpha} \mathbf{S}_{\mathbf{m}_\alpha} - H \sum_{\mathbf{n}_\alpha} S_{\mathbf{n}_\alpha}^Z,$$
(1)

where the spins $\mathbf{S}_{\mathbf{n}_1}$ and $\mathbf{S}_{\mathbf{n}_2}$ belong to the same dimer, $J_0 > 0$ is the parameter of exchange interaction between them, $\mathbf{S}_{\mathbf{m}_1}$ and $\mathbf{S}_{\mathbf{m}_2}$ are the operators of spins that are nearest to the dimer (spins $\mathbf{S}_{\mathbf{n}_1}$ and $\mathbf{S}_{\mathbf{m}_1}$ are related to the first ($\alpha = 1$) sublattice, while spins $\mathbf{S}_{\mathbf{n}_2}$ and $\mathbf{S}_{\mathbf{m}_2}$ to the second one ($\alpha = 2$)), $J > 0$ is the parameter of exchange interaction between the spins of neighbor dimers that belong to the same sublattice, the spin arrangement is determined by vectors $\mathbf{n}$ and $\mathbf{m}$, and $\mathbf{H}$ is the magnetic field parallel to the Z-axis. We consider
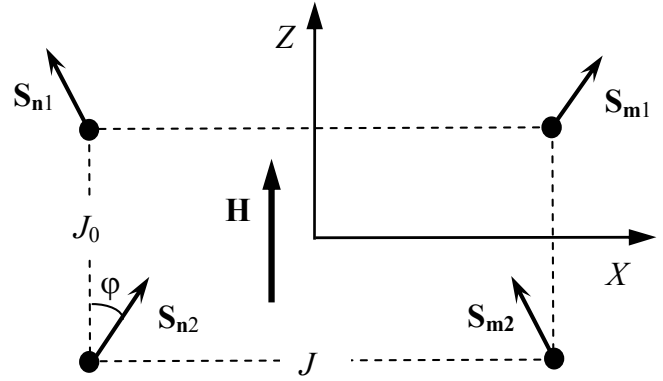

Fig. 1. Spin orientation in neighbor dimers in a magnetic field

further that the magnitude of spins in Eq. (1) $S = 1/2$, and the field and the exchange parameters are given in energy units.

In the self-consistent field approximation, the spins of dimers are supposed to be in the exchange field created by spins in the environment. In this approximation, the Hamiltonian for a dimer can be simplified,

$$\hat{H}_{dim} = J_0 \mathbf{S}_{\mathbf{n}_1} \mathbf{S}_{\mathbf{n}_2} - \mathbf{h}_1 \mathbf{S}_{\mathbf{n}_1} - \mathbf{h}_2 \mathbf{S}_{\mathbf{n}_2} - H(S_{\mathbf{n}_1}^Z + S_{\mathbf{n}_2}^Z),$$
(2)

where $\mathbf{h}_1$ and $\mathbf{h}_2$ are the exchange fields created by the dimer's neighbors and acting on the dimer's spins. The exchange field is

$$\mathbf{h}_\alpha = -Jz\mathbf{s}_\alpha,$$
(3)

where z is the number of the nearest neighbors (e.g., $z = 4$ for $TlCuCl_3$). The quantity $\mathbf{h}_\alpha$ is proportional to the average spin $\mathbf{s}_\alpha$ at the site. In so doing, we assume that the ground state is homogeneous; therefore, the vectors $\mathbf{s}_\alpha$ lose their dependence on $\mathbf{n}$, but retain their dependence on α. Note also that the X-axis, being directed perpendicularly to the Z-axis, lies in the plane formed by the vectors $\mathbf{s}_1$ and $\mathbf{s}_2$.

Hamiltonian (2) makes it evident that, in the framework of the selected approach, only the exchange interaction between different dimers is taken into account self-consistently, whereas the exchange within a dimer is considered precisely. Now, supposing that the sublattices are symmetric, and their spins are canted identically with respect to the magnetic field direction (see Fig. 1), let us write down the evident relations for projections of the vectors $\mathbf{s}_\alpha$:

$$s_1^Z = s_2^Z = s_Z, \quad -s_1^X = s_2^X = s_X.$$
(4)

Accordingly, the projections for exchange fields are also simplified,

$$h_1^Z = h_2^Z = h_Z, \quad -h_1^X = h_2^X = h_X, \qquad (5)$$

and Hamiltonian (2) reads

$$\hat{H}_{\rm dim} = J_0 \mathbf{S}_{\mathbf{n}_1} \mathbf{S}_{\mathbf{n}_2} - h^X S_{\mathbf{n}_1}^X + h^X S_{\mathbf{n}_2}^X -$$

$$-(H + h^Z)(S_{\mathbf{n}_1}^Z + S_{\mathbf{n}_2}^Z). \qquad (6)$$

The characteristic functions of the product $\mathbf{S}_{\mathbf{n}_1}\mathbf{S}_{\mathbf{n}_2}$ are determined by the total pair spin $S_{\rm dim}$ and its projection. For a pair of spins $S = 1/2$, the total spin can be either $S_{\rm dim} = 1$ or $S_{\rm dim} = 0$. Hence, the sought wave functions for such a spin pair look like [5,6]

$$\psi_{\rm dim}^{(1,1)} = |1,1\rangle, \quad \psi_{\rm dim}^{(1,0)} = |1,0\rangle, \quad \psi_{\rm dim}^{(1,-1)} = |1,-1\rangle,$$

$$\psi_{\rm dim}^{(0,0)} = |0,0\rangle, \qquad (7)$$

where the first number describes the total spin, and the second one its projection. It is easy to verify that, if $J_0 > 0$, the ground state of a dimer is the singlet $\psi_{\rm dim}^{(0,0)}$.

If J_0 considerably dominates over J, this singlet is the ground state for Hamiltonian (1) as well. It is evident that, under the action of a large magnetic field, a transition is to be fulfilled into the state with the maximal spin value which is described by the function $\psi_{\rm dim}^{(1,1)}$. Our research aims at describing a phase transition induced by a magnetic field, which occurs from a singlet state through the so-called canted AFM state to the state magnetized as much as possible, and at finding the critical fields of such a transition.

The initial wave functions (7) can be written down in terms of projections of the spin vectors of the first and the second ion of the pair. Then we have

$$\psi_{\rm dim}^{(1,1)} = \left|\frac{1}{2},\frac{1}{2}\right\rangle, \quad \psi_{\rm dim}^{(1,0)} = \frac{1}{\sqrt{2}}\left(\left|\frac{1}{2},-\frac{1}{2}\right\rangle + \left|-\frac{1}{2},\frac{1}{2}\right\rangle\right),$$

$$\psi_{\rm dim}^{(1,-1)} = \left|-\frac{1}{2},-\frac{1}{2}\right\rangle,$$

$$\psi_{\rm dim}^{(0,0)} = \frac{1}{\sqrt{2}}\left(\left|\frac{1}{2},-\frac{1}{2}\right\rangle - \left|-\frac{1}{2},\frac{1}{2}\right\rangle\right). \qquad (8)$$

Using functions (8) as the initial ones, one can easily find that the exchange field h^X in Hamiltonian (6) mixes the

states $\psi_{\rm dim}^{(1,1)}$ and $\psi_{\rm dim}^{(0,0)}$, so that the wave functions of a dimer are

$$\psi_{\rm dim}^{(1)} = C_1 \psi_{\rm dim}^{(1,1)} + C_2 \psi_{\rm dim}^{(0,0)}, \ \ \psi_{\rm dim}^{(2)} = -C_2 \psi_{\rm dim}^{(1,1)} + C_1 \psi_{\rm dim}^{(0,0)},$$

$$\psi_{\rm dim}^{(3)} \equiv \psi_{\rm dim}^{(1,0)}, \quad \psi_{\rm dim}^{(4)} \equiv \psi_{\rm dim}^{(1,-1)}, \qquad (9)$$

where the constants C_1 and C_2 are interconnected by the condition $|C_1|^2 + |C_2|^2 = 1$. The third and the fourth function in expressions (9) correspond to nonperturbed states of the dimer. If the AFM interaction prevails, just these states have positive energies and lie above the ground state.

The first and the second state in expressions (9) are superpositions of two initial dimer states with different multiplicities. Thereby, we come to the known statement that the eigenstate of the antiferromagnet is characterized by an indefinite multiplicity. The corresponding mixing is caused by the action of an exchange AFM field between the spins of different dimers. The wave functions (9) correspond to the AFM phase with nonzero average projections of every spin. One should bear in mind that making use of the Néel state as the ground one prohibits from the indicated mixing of states and, as a consequence, gives rise to the absence of a phase transition from the quantum-disordered (singlet) phase into a magnetically ordered one.

In the case where the coefficients C_1 and C_2 equal their limiting values (0 or 1), the system is either in the singlet state (if the field is absent) or in the ferromagnetic state (if the field is very large). In the general case, those coefficients depend on H.

3. Equation of State

Let us determine the energy of the ground state of exchange-coupled dimers. If the interaction between dimer's spins is taken into account precisely, and the interaction between spins of different dimers is made allowance for in the mean-field approximation, the energy of the system per dimer can be written down in the following form:

$$E_{\rm dim}^{\rm (gr)} = J_0 \langle \mathbf{S}_1 \mathbf{S}_2 \rangle + 2Jz\mathbf{s}_1\mathbf{s}_2 - H(s_1^Z + s_2^Z), \qquad (10)$$

where the notation $\langle \ldots \rangle$ stands for the quantum-mechanical averaging, and $\mathbf{s}_\alpha$ means the quantum-mechanical average for spin vectors.

In the XZ-coordinate system introduced above (see Fig. 1), the quantities $\langle \mathbf{S}_1 \mathbf{S}_2 \rangle$ and $\mathbf{s}_\alpha$ that appear in

Eq. (10) can be calculated making use of the first function from set (9). Then, we have

$$\langle \mathbf{S}_1 \mathbf{S}_2 \rangle = \left\langle \psi^{(1)} \middle| \mathbf{S}_1 \mathbf{S}_2 \middle| \psi^{(1)} \right\rangle = \frac{1}{4} C_1^2 - \frac{3}{4} C_2^2, \qquad (11)$$

$$s_X = - \left\langle \psi^{(1)} \middle| s_1^X \middle| \psi^{(1)} \right\rangle = \left\langle \psi^{(1)} \middle| s_2^X \middle| \psi^{(1)} \right\rangle = \frac{1}{2} C_1 C_2, \qquad (12)$$

$$s_Z = \left\langle \psi^{(1)} \middle| s_1^Z \middle| \psi^{(1)} \right\rangle = \left\langle \psi^{(1)} \middle| s_2^Z \middle| \psi^{(1)} \right\rangle = \frac{1}{2} C_1^2. \qquad (13)$$

Taking advantage of expressions (11)–(13), we can write down the energy of the ground state as

$$E_{\mathrm{dim}}^{(\mathrm{gr})}(C_1) = -\frac{3}{4} J_0 + \left(J_0 - \frac{1}{2} Jz - H \right) C_1^2 + Jz C_1^4. \qquad (14)$$

In this state, the energy must be minimal [10, 11]. In other words, the derivative $dE_{\mathrm{dim}}^{(\mathrm{gr})}/dC_1 = 0$ in the ground state. Differentiating expression (14), we arrive at the equation to find the coefficient C_1:

$$2\left(J_0 - \frac{1}{2} Jz - H + 2Jz C_1^2 \right) C_1 = 0. \qquad (15)$$

One should bear in mind that $C_1 \in [0,1]$, and this variational parameter can reach its limiting values.

4. Phase Transitions

Equation (15) gives rise to a result that is impossible for a classic or quasiclassical magnet: a dimerized spin system can be in a singlet state, which corresponds to the condition $C_1 = 0$, even in a finite magnetic field. In turn, it testifies that the wave function $\psi_{\mathrm{dim}}^{(1)}$ (see Eq. (9)) remains constant, i.e. it is equal to zero. The energy of this state evidently depends neither on the field H nor on the parameter J: $E_{\mathrm{dim}}^{(0,0)} = -3J_0/4$. In the AFM state with two sublattices, on the contrary, the coefficient C_1 depends on H:

$$C_1 = \sqrt{\frac{H + (1/2)Jz - J_0}{2Jz}}. \qquad (16)$$

In this state with $C_1 \neq 0$, the dimer functions are mixed. This circumstance results in the dependence of averages (11)–(13) on the model parameters and – it is the key moment! – on the field magnitude. Dimer ions get polarized, and they acquire a magnetic moment. Therefore, it should be emphasized that the ordering, in which $(C_1, C_2) \neq 0$, must be defined as a quantum

AFM state. This ordering essentially differs from that of the Néel type, in which spin states with different multiplicities do not mix together. This is the key difference of dimerized magnets from the van Vleck ones. The latter are also singlet; the corresponding spin states mix together too, but within a single spin multiplet.

The energy of this AFM state is

$$E_{\mathrm{dim}}^{(\mathrm{AFM})} = -\frac{3J_0 Jz + [J_0 - (1/2)Jz - H]^2}{4Jz}. \qquad (17)$$

One can see that the field reduces the energy of an "AFM-ordered" dimer. It is also evident that the AFM phase can be formed only at fields larger than a definite critical field

$$H_{\mathrm{cr}}^{(1)} = J_0 - \frac{1}{2} Jz. \qquad (18)$$

The magnitude of the latter is determined by the condition that coefficient (16) must be real-valued.

When the field reaches the $H_{\mathrm{cr}}^{(1)}$-value, the singlet state smoothly—rather than in a jump-like manner— transforms into an AFM state, where sublattices' spins are canted with respect to the external field direction. A further field increase brings the system into a ferromagnetic (paramagnetic) state, in which the coefficient C_1 reaches another limiting value of 1. In so doing, either the Z-projections of both dimer's spins become maximal or the sublattices' spins become parallel to one another. The energy of this state is

$$E_{\mathrm{dim}}^{(\mathrm{PM})} = \frac{1}{4} J_0 + \frac{1}{2} Jz - H. \qquad (19)$$

The critical field of the transition from canted AFM states into the paramagnetic one is determined by the expression

$$H_{\mathrm{cr}}^{(2)} = J_0 + \frac{3}{2} Jz. \qquad (20)$$

As is seen from Eqs. (18) and (20), $H_{\mathrm{cr}}^{(2)} > H_{\mathrm{cr}}^{(1)}$.

Hence, the simple model of a system consisting of dimers magnetically biased due to the exchange interaction demonstrates that it can naturally undergo a magnetic QPT from the singlet (quantum-disordered) state into a quantum AFM state. The latter is essentially quantum owing to a reduction of the "site"-spin (to be more exact, dimer-spin) magnitude, whereas there is no such reduction for a spin $S = 1/2$ system in the quasi-classical approximation. The corresponding QPT occurs smoothly, i.e. it is a phase transition of the second kind.

5. Conditions for Exchange Interaction Parameters

Let us determine the relations between the model constants, provided that the initial ground state of every dimer spin is singlet and the application of an external magnetic field induces the QPT into the AFM state. For this purpose, in addition to the states considered above, we must also take into account that the Néel AFM phase with antiparallel dimer spins can be formed.

In the classical approximation, the absolute value of spins in the sublattices is $|\mathbf{s}_\alpha| = 1/2$. The magnetic field changes their direction only. In this case, the equilibrium orientation of spin vectors is determined by minimizing the energy

$$E_{\text{Néel}}^{(\text{AFM})} = (J_0 + 2Jz)\mathbf{s}_1\mathbf{s}_2 - H(s_1^Z + s_2^Z). \qquad (21)$$

The average spins in both sublattices become canted identically or symmetrically with respect to the field vector. The angle φ between the spin vector and the field direction is determined by the expression

$$\cos\varphi = \frac{H}{J_0 + 2Jz}. \qquad (22)$$

Taking Eq. (22) into account, it is easy to calculate the energy of the Néel phase,

$$E_{\text{Néel}}^{(\text{AFM})} = -\frac{1}{4}(J_0 + 2Jz) - \frac{H^2}{2(J_0 + 2Jz)}, \qquad (23)$$

the latter can exist up to the field

$$H_{\text{cr}}^{(\text{AFM}\to\text{FM})} = J_0 + 2Jz \qquad (24)$$

in the quasi-classical approach. Hence, the expressions obtained allow one to determine such parameters, which would ensure that the singlet phase is the ground state of the model at $H = 0$. For this to be true, the energy of the singlet phase should evidently be lower than that of the Néel phase. By comparing them, we find a simple and rather clear inequality

$$J_0 > Jz. \qquad (25)$$

In essence, it defines a condition for the quantum paramagnetic state to be formed in a dimerized antiferromagnet as the ground one.

In order that the phase transition into a quantum AFM state with a reduced average spin of ions occurs in a magnetic field, it is necessary that the energy of the singlet phase remain lower than the energy of the Néel

phase up to the point $H = H_{\text{cr}}^{(1)}$. In other words, the inequality

$$-\frac{3}{4}J_0 < -\frac{1}{4}(J_0 + 2Jz) - \frac{(H_{\text{cr}}^{(1)})^2}{2(J_0 + 2Jz)} \qquad (26)$$

must remain valid. If definition (18) is taken into account, inequality (26) is reduced to the relation

$$J_0 > \frac{9}{8}Jz \qquad (27)$$

which turns out to be more strict than inequality (25). Therefore, condition (27) somewhat confines the range of existence of the singlet phase as the ground one in the model of spin dimers "magnetically biased" by the exchange field.

We now consider whether there can be a transition from the quantum AFM into the classic AFM phase. For this purpose, let us write down the equality $E_{\text{dim}}^{(\text{AFM})} = E_{\text{Néel}}^{(\text{AFM})}$ which implies that such a phase transition occurs in the field

$$H_{\text{cr}}^{(3)} = J_0 + \frac{3}{2}Jz - \frac{(Jz)^2}{J_0}\left(1 + \sqrt{1 + \frac{J_0}{2Jz}}\right). \qquad (28)$$

In the course of this transition, the absolute value of the average spin changes in a step-wise fashion, i.e. this transition turns out to be of the first kind.

However, it should be noted that both condition (27) and expression (28) are not strict enough, since they were derived making use of inherently different methods of calculation of the ground state energy. But despite that, we may make some remarks concerning the results of work [6], where the isotropic exchange model (1) was also analyzed, but no possibility was found for a phase transition of the first kind to occur. On the contrary, we do see that this model provides such a possibility. Therefore, the corresponding transformation in the field $H_{\text{cr}}^{(3)}$ should be considered by analogy with isostructural phase transitions. For the first time, phase transitions, similar to isostructural ones, between two classical AFM phases were described by Bar'yakhtar et al. [16]. In our case, a phase transition of the first kind between a singlet and a Néel phase should be observed, provided that $Jz < J_0 < 9Jz/8$. The fulfillment of this condition in real systems, as well as the applications of the isotropic model to them, requires special researches.

6. Properties of Quantum AFM Phase

Let us analyze how the properties of the system of exchange-coupled dimers change at dimer magnetization. In the singlet phase, the magnetization is absent; in other words, the magnetic susceptibility is absent at $H < H_{\mathrm{cr}}^{(1)}$: $\chi_{\mathrm{dim}}^{(0,0)}(H < H_{\mathrm{cr}}^{(1)}) = 0$.

At $H \geq H_{\mathrm{cr}}^{(1)}$, there emerges a quantum AFM phase. As follows from expressions (13) and (16), the Z-projection of a spin in it,

$$s^Z(H) = \frac{H - H_{\mathrm{cr}}^{(1)}}{2(H_{\mathrm{cr}}^{(2)} - H_{\mathrm{cr}}^{(1)})}, \tag{29}$$

linearly depends on the field, which excellently agrees with experimental data [5,6].

According to expression (29), if $H > H_{\mathrm{cr}}^{(1)}$, there appears a Z-projection linearly growing with the field and reaching its evident limiting value $s^Z = 1/2$ at the point $H = H_{\mathrm{cr}}^{(2)}$, if no phase transition into the Néel phase occurs. In Fig. 2, an example of the relevant dependence is plotted, which corresponds to the ratio $H_{\mathrm{cr}}^{(2)}/H_{\mathrm{cr}}^{(1)} = 3$.

Another, X-projection of the average spin also arises at the field $H = H_{\mathrm{cr}}^{(1)}$, and it also starts from the zero value. Its field dependence is described by the expression

$$s^X(H) = \frac{\sqrt{(H - H_{\mathrm{cr}}^{(1)})(H_{\mathrm{cr}}^{(2)} - H)}}{4(H_{\mathrm{cr}}^{(2)} - H_{\mathrm{cr}}^{(1)})}. \tag{30}$$

One can see from this formula that the X-projection vanishes at $H = H_{\mathrm{cr}}^{(2)}$. This phenomenon stems from the phase transition of the spin-flip type (the dependence $s^X(H)$ is plotted in Fig. 2).

From expression (30), it also follows that, in the upper vicinity of $H_{\mathrm{cr}}^{(1)}$ ($H > H_{\mathrm{cr}}^{(1)}, H \to H_{\mathrm{cr}}^{(1)}$), the X-projection of the spin is approximately determined by the formula

$$s^X(H) \approx \frac{1}{2}\sqrt{\frac{H - H_{\mathrm{cr}}^{(1)}}{2Jz}}, \tag{31}$$

which is similar to the mean-field critical dependence.

A comparison between expressions (29) and (30) demonstrates that, if $H \to H_{\mathrm{cr}}^{(1)}$ from above, the spins in sublattices are oriented normally to the field, with the spins of either sublattice being practically antiparallel to the spins of the other sublattice. In other words, at the point $H = H_{\mathrm{cr}}^{(1)}$ of the first phase transition, there emerges a spontaneous spin polarization in the crystal,
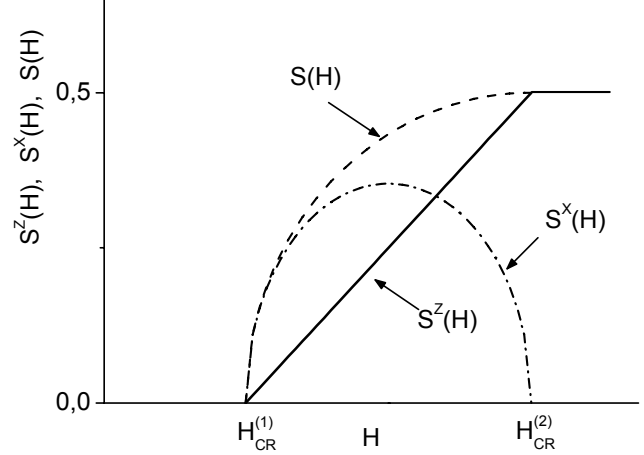

Fig. 2. Field dependences of the projections $s^Z(H)$ and $s^X(H)$ and the absolute value $s(H)$ of spins in the sublattices

which can be described by an AFM vector perpendicular to the **H**-direction. The field growth is accompanied by the growth of the spin polarization; the cant of the sublattice spin vectors with respect to the magnetic field also increases.

Expression (29) gives rise to a strange conclusion: in spite of the spin reduction, the magnetic susceptibility of the quantum AFM phase in the field range $H_{\mathrm{cr}}^{(1)} < H < H_{\mathrm{cr}}^{(2)}$ – or $H_{\mathrm{cr}}^{(1)} < H < H_{\mathrm{cr}}^{(3)}$, if the phase transition into the Néel phase occurs – does not depend on H, namely,

$$\chi_{\mathrm{dim}}^{(\mathrm{AFM})} = \frac{ds^Z(H)}{dH} = \frac{1}{4Jz}. \tag{32}$$

From formulas (12) and (13), we also find another, rather unexpected, relation for spin projections:

$$(s^X)^2 + (s^Z)^2 = \frac{1}{2}s^Z. \tag{33}$$

It shows that, in the field range $H_{\mathrm{cr}}^{(1)} < H < H_{\mathrm{cr}}^{(2)}$ (or $H_{\mathrm{cr}}^{(1)} < H < H_{\mathrm{cr}}^{(3)}$), not only the projections of the spin, but its absolute value as well, depend on H in a critical manner. In particular, for the latter,

$$|\mathbf{s}| \equiv s(H) = \sqrt{(s^X)^2 + (s^Z)^2} = \frac{1}{2}\sqrt{\frac{H - H_{\mathrm{cr}}^{(1)}}{H_{\mathrm{cr}}^{(2)} - H_{\mathrm{cr}}^{(1)}}}. \tag{34}$$

The last expression testifies that the absolute value s of the average spin in the quantum AFM phase depends on the field H and varies from 0 to 1/2. Hence, the spin in this quantum phase is also reduced, being smaller than its maximal value (the plot of the $s(H)$-dependence is exhibited in Fig. 2).

7. Conclusions

Our calculations allow a conclusion to be drawn that the induced transition in a dimerized antiferromagnet from the singlet state into the quantum AFM one occurs spontaneously, through the spin polarization of dimers. The magnitude of this polarization depends on the magnetic field. The influence of the magnetic field is caused by the emergence of average spins–absent before–of sublattices' ions and their growth in the increasing field. The growing field also induces a spin cant with respect to the magnetic field direction. In spite of a strong non-linearity of the process, the magnetic susceptibility of the quantum AFM phase remains independent of the field.

It is also shown that the standard approach of the Landau theory of phase transitions is applicable to the description of the magnetic QPT from the singlet state into the quantum two-sublattice AFM state. In this case, the ground state and its characteristics are determined by minimizing the energy. However, the temperature effects in the system and the corresponding calculations of the free energy require a special study. Among the conclusions of this work is the proved, in fact, absence of the necessity to engage the ideas and methods of the BEC theory for the description of such magnetic QPTs.

At last, we obtained that making allowance for only isotropic exchange interactions in the model enables the latter to describe the isostructural phase transition of the first kind from the quantum AFM phase into the Néel one. For the TlCuCl$_3$ compound, such a phase transition is not observed. It may mean, first of all, that a model for studying the properties of this dimerized magnetic system cannot include isotropic interactions only.

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МАГНІТНІ КВАНТОВІ ФАЗОВІ ПЕРЕХОДИ
В ДИМЕРИЗОВАНИХ АНТИФЕРОМАГНЕТИКАХ

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Р е з ю м е

Здійснено теоретичний опис індукованого магнітним полем квантового фазового переходу в системі обмінно зв'язаних спінів $S = 1/2$, що формують димери з синглетним основним станом і зв'язані між собою антиферомагнітно. Показано, що під дією магнітного поля критичним чином відбувається перехід у квантову антиферомагнітну фазу. Параметром порядку за такого фазового переходу є спонтанно виникаюча спінова поляризація іонів димера, що відносяться до різних підґраток. Величина модуля середніх спінів підґраток у квантовій антиферомагнітній фазі залежить від величини магнітного поля, а її магнітна сприйнятливість залишається сталою. Показано, що у наближенні ізотропних спін-спінових взаємодій є можливим ізоструктурний фазовий перехід між квантовою антиферомагнітною та неєлівською фазами.