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Alternating Heisenberg $s=\frac{1}{2}$ spin chain: ferromagnetism versus antiferromagnetism

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Spin ladder systems containing $CuCl_3$ and $CuBr_3$ show very interesting magnetic behaviour, which are alternate Heisenberg chains with ferromagnetic and antiferromagnetic interactions. Since these systems contain triangles of magnetic centers, we developed a model based on non-interacting triangular units, which may be used for interpretation of magnetic measurements of these systems in some extreme limits. We took the Heisenberg model type interactions for magnetic center and used different types of exchange coupling between the neighbours. The results obtained from our approximations may be used in order to interpret the experimental magnetic measurements in $KCuCl_3$ and $TlCuCl_3$ complexes and for the singlet-triplet transition temperature.

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1. Introduction

Spin ladder systems containing $CuCl_3$ and $CuBr_3$ show very interesting magnetic behaviour, which is alternate Heisenberg chains with ferromagnetic and antiferromagnetic (F-AF) interactions. It has been suggested that alternating F-AF Heisenberg chains with S=1/2 behave like antiferromagnetic Heisenberg chains with S=1, when the ferromagnetic exchange is dominant[1]. The energy spectrum then has an energy gap between the ground state and the first excited state. The system of $KCuCl_3$ and $TlCuCl_3$ both have double chain structure shown in Figure-1.a of Ref [2]. In these compounds all of the octahedral arrangements are distorted due to the Jahn-Teller effect. Experimental studies for these compounds suggest that the interactions between Cu ions may be different in different directions as shown in Figure-1.b of Ref. [2]. Although the experiments for these compounds have been carried under high magnetic field, we will concentrate on nearly non interacting triangular units of spins. Once understanding the magnetic behaviour of these units, we are expecting to be in a position to attact the high field case. It should be noted that this problem will be studied in three different situations. In the first case we will consider the isotropic limit where all the interactions are ferromagnetic. In the second case the same problem will be studied with antiferromagnetic interactions and in the final stage we will let the interaction to be mixed(ferromagnetic and antiferromagnetic interactions). So firstly we will take the ferromagnetic case and use a suitable model in order to understand the magnetic behaviour and the effect of magnetic field to these triangular units.

2. Model and calculation details

2.1 The model Hamiltonian for isotropic ferromagnetic interaction

In our study we used the Heisenberg exchange type interactions where the Hamiltonian is

$$H = -J \sum_{i\,j} \vec{S}_i . \vec{S}_j \qquad (1)$$

here the J is exchange coupling constant and $\vec{S}_i$, $\vec{S}_j$ are nearest neighbour spins. Since the system is ferromagnetic the coupling constant J must be positive. For a trinuclear triangular arrangement of spins (Fig. 1) this equations can be rewritten as

$$H = -J\left(\vec{S}_1.\vec{S}_2 + \vec{S}_2.\vec{S}_3 + \vec{S}_1.\vec{S}_3\right) \qquad (2)$$

In terms of spin components or more precisely with the spin operators this equation can be organised as

$$H = -\frac{J}{2}\left(S_1^+ S_2^- + S_1^- S_2^+ + S_2^+ S_3^- + S_2^- S_3^+ + S_1^+ S_3^- + S_1^- S_3^+\right) - J\left(S_{1z}S_{2z} + S_{2z}S_{3z} + S_{1z}S_{3z}\right) \qquad (3)$$

Diagonalizing this Hamiltonian in the base of spin functions one can set the Hamiltonian matris as

$$H = J\begin{bmatrix} -3/4 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -3/4 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1/4 & -1/2 & -1/2 & 0 & 0 & 0 \\ 0 & 0 & -1/2 & 1/4 & -1/2 & 0 & 0 & 0 \\ 0 & 0 & -1/2 & -1/2 & 1/4 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1/4 & -1/2 & -1/2 \\ 0 & 0 & 0 & 0 & 0 & -1/2 & 1/4 & -1/2 \\ 0 & 0 & 0 & 0 & 0 & -1/2 & -1/2 & 1/4 \end{bmatrix} \quad (4)$$

which has –0.75J and 0.75J eigenvalues. Each eigenvalue has four different eigenfunctions. Using these eigenfunctions one can easily calculate the first order Zeeman effect with the perturbeted Hamiltonian,

$$H' = g\,\mu_B\left(\hat{S}_{1z}+\hat{S}_{2z}+\hat{S}_{3z}\right)H \quad (5)$$

Using this Hamiltonian one can get first order Zeeman contributions as

$$W_1^{(1)}=(3/2)g\,\mu_B \quad , \quad W_2^{(1)}=(-3/2)g\,\mu_B \quad , W_3^{(1)}=(1/2)g\,\mu_B \quad , W_4^{(1)}=(-1/2)g\,\mu_B$$
$$W_5^{(1)}=(1/2)g\,\mu_B \quad , \quad W_6^{(1)}=(1/2)g\,\mu_B \quad , W_7^{(1)}=(-1/2)g\,\mu_B \quad , W_8^{(1)}=(-1/2)g\,\mu_B \quad (6)$$

Using these energies one may write the magnetic susceptibility as

$$\chi = \frac{N\sum_i W_i^{(1)^2}\,\exp\left(-W_i^{(0)}/kT\right)}{kT\sum_i \exp\left(-W_i^{(0)}/kT\right)} \quad (7)$$

The variation of χ as a function of temperature is pictured in Fig. 2.

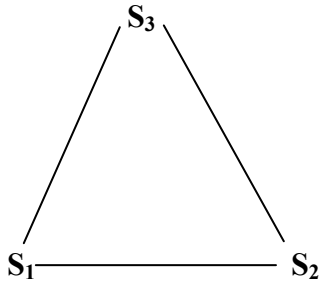

Fig. 1. Trinuclear triangular arrangement of spins.

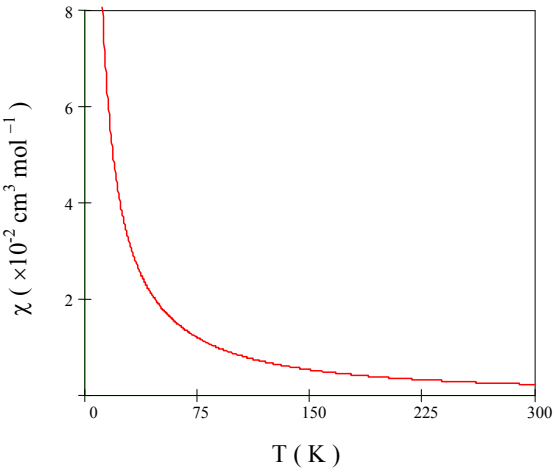

Fig. 2. The variation of magnetic susceptibility as a function of temperature at $\alpha=1$ *and* $J=100\,cm^{-1}$.

Using this susceptibility we can express the effective magnetic moment as

$$\mu = (\chi\,T)^{1/2} \quad (8)$$

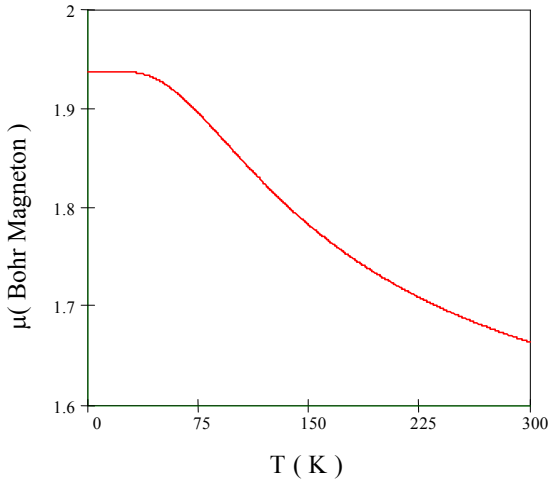

Fig. 3. The variation of magnetic moment as a function of temperature at $\alpha=1$ *and* $J=100\,cm^{-1}$.

The variation of μ as a function of temperature shows that this system is a ferromagnetic one, and we get the saturation magnetization at T=0 shown in Fig. 3.

This is the expected result which is generally observed in many chemical compounds Ref [3,4,5,6,7].

2.2 İsotropic antiferromagnetic interaction

The same Hamiltonian(Equation(2)) can be used for the antiferromagnetic interactions with J< 0. In this situation the diagonalized Hamiltonian leading to two fourfold degenerate energy.

$$H = -J \begin{bmatrix} -3/4 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -3/4 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1/4 & -1/2 & -1/2 & 0 & 0 & 0 \\ 0 & 0 & -1/2 & 1/4 & -1/2 & 0 & 0 & 0 \\ 0 & 0 & -1/2 & -1/2 & 1/4 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1/4 & -1/2 & -1/2 \\ 0 & 0 & 0 & 0 & 0 & -1/2 & 1/4 & -1/2 \\ 0 & 0 & 0 & 0 & 0 & -1/2 & -1/2 & 1/4 \end{bmatrix} \quad (9)$$

Performing the same calculation which is done for the ferromagnetic case, one can get the susceptibility as

$$\chi = \frac{N \sum_i W_i^{(1)^2} \exp\left(-W_i^{(0)}/kT\right)}{kT \sum_i \exp\left(-W_i^{(0)}/kT\right)} \quad (10)$$

and the magnetic moment

$$\mu = (\chi T)^{1/2} \quad (11)$$

The variation of the magnetic moment as a function of temperature is shown in Fig. 4. The moment approaches to the value of 1/2. This is the expected value for a "triangular" arrangement of any spin 1/2 antiferromagnetic system, when the magnetic moments force to arrange in the z direction.

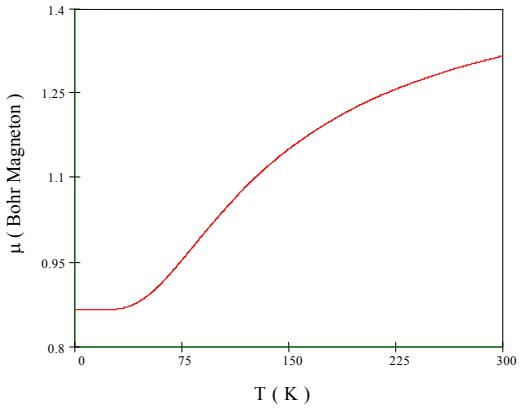

Fig. 4. The variation of magnetic moment as a function of temperature at $\alpha = 1$ *and* $J = -100\,cm^{-1}$.

2.3 Ferromagnetic-antiferromagnetic mixed interaction

For many chemical compounds especially the systems we have been considering, the interactions may be different in nature. This may be due to the fact that each magnetic metal ion (Cu^{2+} in our systems) has a different chemical environment. This may lead to different kinds of exchange interactions. In this part of our study we take two different kinds of exchange interactions for our model Hamiltonian and try to get some comprehensive results which may be used to interpret the experimental data. The Hamiltonian for this situation is

$$H = -J_1 \vec{S}_1 . \vec{S}_2 - J_2 \left(\vec{S}_1 . \vec{S}_3 + \vec{S}_2 . \vec{S}_3 \right). \quad (12)$$

Defining J_1 / J_2 as α one can rewrite this Hamiltonian as

$$H = -J_2 \left(\alpha \vec{S}_1 . \vec{S}_2 + \vec{S}_1 . \vec{S}_3 + \vec{S}_2 . \vec{S}_3 \right). \quad (13)$$

Using the experimental value for J_2=42.4 and α = 0.55 one can obtain the χ as

$$\chi = \frac{N g^2 \beta^2}{kT} \left[\frac{5\exp(22.26/kT) + (1/2)\exp(-31.8/kT) + (1/2)\exp(-12.72/kT)}{4\exp(22.26/kT) + 2\exp(-31.8/kT) + 2\exp(-12.72/kT)} \right] \quad (14)$$

which lead to a magnetic moment as

$$\mu = (\chi T)^{1/2} \quad (15)$$

Their dependence on T is shown in Fig. 5 and Fig. 6, respectively.

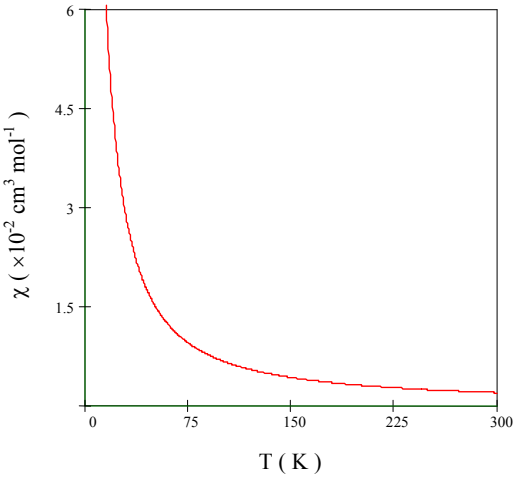

Fig. 5. The variation of magnetic susceptibility as a function of temperature at $\alpha = 0.55$ *and* $J = 42.4\,cm^{-1}$.

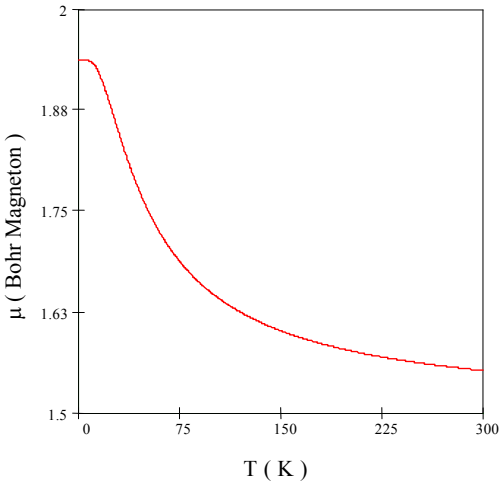

Fig. 6. The variation of magnetic moment as a function of temperature at $\alpha = 0.55$ *and* $J = 42.4\,cm^{-1}$.

Experimental studies suggest that under the high field, these system shows a singlet-triplet transition at around T=40 K. We take this particular temperature and look at the variation of the effective magnetic moment as a function of α. It is quite clear from this picture that the result obtained by us is in fairly go agreement experimental results (Fig. 7). Three particular value of α need to a few words: These are α=0, α=1 and α= -1. α=0 means two bonds are broken and just one strong interaction is active and that is ferromagnetic. Since this interaction is ferromagnetic, we should get spin 1 at this particular value of α, which is obtained in our model too. At two other extremes the calculations are also not far away from the experimental results. Making all bonds ferromagnetic and isotropic (α=1). We should have a ferromagnetic alignment of spins and maximum magnetic moment which is clear from our calculations.

α= -1 means two bonds are antiferromagnetic and one bond is ferromagnetic. In this case one must get spin 1/2. So the results obtained from our approximations may be used in order to interpret the experimental magnetic measurements in $KCuCl_3$ and $TlCuCl_3$ complexes.

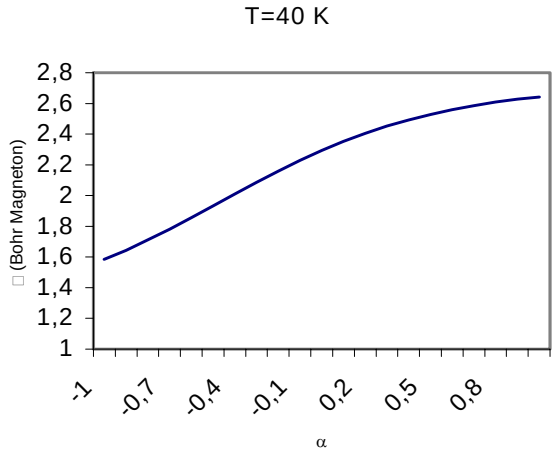

Fig. 7. The variation of magnetic moment as a function of α at T=40 K.

3. Results and discussion

Experimental results for the spin ladder systems containing $CuCl_3$ and $CuBr_3$ indicate that under high magnetic field these systems undergo a singlet-triplet transition at around T=40 K. This is a kind of phase transition from an antiferromagnetic arrangement to a ferromagnetic one. Since the environments of all magnetic centers are different, the magnetic interactions between these centers can not be the same. It may differ in magnitude and sign. In this study we firstly concentrated on isotropic cases and then tried to include unisotropic interactions. The results obtained for isotropic cases (both ferromagnetic and antiferromagnetic) are in a good agreement with experiments for interacting three nuclear triangular arrangements. The reason for getting a magnetic moment different from zero in the antiferromagnetic case is the existence of odd number of interacting bonds. In such a case not all neighbours can be antiparallel. This problem can be understood as a spin frustration phenomenon. Spin frustration in many different systems has been studied recently Ref [8,9,10,11,12]. A simple picture of a frustration in a triangular unit may be explained as follows: In Fig. 1 we are taking the triangular unit as an example. For an antiferromagnetic arrangement the sum of the three spins ($\vec{S}_1+\vec{S}_2+\vec{S}_3=0$) must give zero spin. If we make two of these spins, as show in Fig. 8 antiparallel to each other the third, spin can not be made antiparallel to the first two spins. This is the simple picture of frustration. The frustration can be seen in many geometry for antiferromagnetic interactions. In such systems the ground state spin arrangements are generally degenerate and have at least two spin components (not arranged in a unique direction). For example in order to get a zero spin from the sum of three spins in a triangular unit, a possible spin arrangement should be as in Fig. 9, where the rotational degeneracy remain. This kind of arrangement can not be deduced in a simple manner. A possible way may be a neutron diffuse scattering. In mixed interactions (ferromagnetic and antiferromagnetic) we mostly concentrated on a particular temperature where the singlet-triplet transition is observed (T= 40 K); and let the relative bond strenght to vary. We saw that at the limits of α=1 and α= -1 the systems behave as ferromagnet and antiferromagnet, respectively. This result may be used to interpret the experimental results where the singlet-triplet transition occurs. One final comment should be pointed out that our approach may be used near the transition temperature.

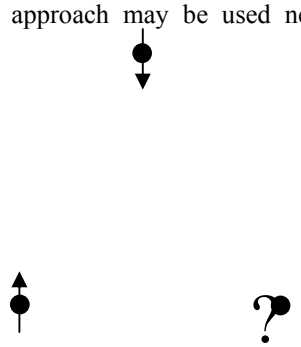

Fig. 8. This figure is the simple picture of frustration.

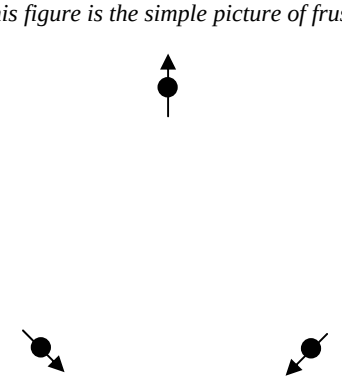

Fig. 9. A possibility of spin arrangements in order to get a zero spin from the sum of three spins in a triangular unit.

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