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Magnetic properties of a $S = 1/2$ zigzag spin chain compound $(\text{N}_2\text{H}_5)\text{CuCl}_3$

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Abstract

We present a theoretical and experimental study of a quasi-one-dimensional zigzag antiferromagnet $(\text{N}_2\text{H}_5)\text{CuCl}_3$, which can be viewed as weakly coupled Heisenberg chains with a frustrated interaction. We first discuss generic features of the magnetic properties of the zigzag spin chain between the nearly single chain case and the nearly double chain case, on the basis of the finite temperature density-matrix renormalization group (DMRG) calculations. We next show the experimental results for the magnetic susceptibility and the high-field magnetization of a single crystal of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ above the Néel temperature $T_N = 1.55$ K. By comparing the experimental data with the DMRG results carefully, we finally obtain the ratio of the nearest and next-nearest exchange couplings as $J_1/J_2 = 0.25$ with $J_2/k_B = 16.3$ K. We also investigate the three-dimensional (3D) coupling J' effect by using mean-field theory combined with the DMRG calculations. The estimated value $J' \sim 0.04J_2$ supports the one-dimensionality of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ above T_N .

1. Introduction

Frustration effects in low-dimensional antiferromagnetic (AF) quantum spin systems have attracted much attention because of their peculiar behaviour in low-energy physics. The quantum effect and the geometrical frustration cooperatively induce a strong fluctuation, which often gives rise to various non-magnetic ground states and quantum phase transitions [1–5].

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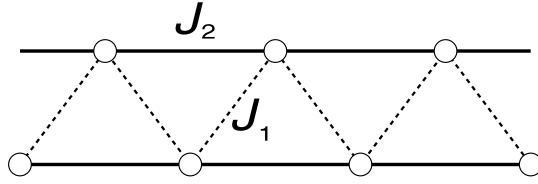


Figure 1. The zigzag spin chain. The dashed lines and solid lines denote the nearest neighbour coupling J_1 and the next-nearest neighbour coupling J_2 respectively.

A typical example of such frustrated low-dimensional magnets is the $S = 1/2$ AF zigzag chain (see figure 1). The Hamiltonian of the zigzag spin chain is given by

$$\mathcal{H} = \sum_i [J_1 \vec{S}_i \cdot \vec{S}_{i+1} + J_2 \vec{S}_i \cdot \vec{S}_{i+2}] - g\mu_B H \sum_i S_i^z, \quad (1)$$

where \vec{S} is the $S = 1/2$ spin operator, g is the Landé g factor and μ_B is the Bohr magneton. J_1 and J_2 denote the nearest neighbour and the next-nearest neighbour coupling constants respectively, and H is the applied magnetic field. Intensive theoretical studies have shown that the zigzag chain has a rich phase structure at zero magnetic field. When $J_2 = 0$, the system is equivalent to the Heisenberg chain having gapless excitation [6]. Increasing J_2 , a quantum phase transition from the critical spin liquid phase to the gapped dimer phase [2, 7] occurs at $J_2/J_1 = 0.2411$ [8]. This gapped dimer phase is expected to extend up to the limit $J_2/J_1 = \infty$ [9], which corresponds to the two decoupled Heisenberg chains. Here, it should be remarked that the dimer gap Δ for $J_1 \ll J_2$ is exponentially small: $\Delta \sim \exp(-\text{constant} \times J_2/J_1)$ [9].

The theoretical studies mentioned above have stimulated experimental studies of the zigzag spin chain. In fact, the magnetic properties of SrCuO_2 [10] and $\text{Cu}[2-(2\text{-aminomethyl})\text{pyridine}]$ [11] were investigated recently. For the latter case, the nearly gapless behaviour of the magnetic susceptibility was observed for a powder sample, and then the ratio of the couplings of $\text{Cu}[2-(2\text{-aminomethyl})\text{pyridine}]$ is estimated as $J_2/J_1 \sim 0.2$ [11]. In this paper, we focus on another type of the zigzag compound $(\text{N}_2\text{H}_5)\text{CuCl}_3$ [12], which is interestingly expected to have the coupling constant $J_1 \ll J_2$ [13]. Since $(\text{N}_2\text{H}_5)\text{CuCl}_3$ can be synthesized as a single crystal, we can precisely compare its magnetic properties with numerical calculations.

An important point on the compound is that its magnetic property is very similar to the one in the single Heisenberg chain limit, although it has the double chain structure. Thus how we can distinguish the magnetic behaviour of the zigzag chains between $J_1 \gg J_2$ and $J_1 \ll J_2$ is an important problem from both theoretical and experimental views. In what follows, we study the magnetic properties of a single crystal sample of $(\text{N}_2\text{H}_5)\text{CuCl}_3$, on the basis of the precise comparison between the numerical results obtained by the finite temperature density-matrix renormalization group (DMRG) method [14, 15] and the experimental measurements of the magnetic susceptibility and the high-field magnetization. In addition, we remark the effect of the three-dimensional (3D) coupling⁶ (J') on $(\text{N}_2\text{H}_5)\text{CuCl}_3$; we apply the mean-field theory combined with the finite temperature DMRG to verify the one-dimensionality of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ [16, 17].

In section 2, we describe the finite temperature DMRG method and the fundamental aspects of the single crystal of $(\text{N}_2\text{H}_5)\text{CuCl}_3$. In section 3, we show the DMRG results for the

⁶ The 3D coupling noted here is usually called ‘inter-chain coupling’. However this naming can be confused with ‘inter-chain coupling’ originating from the double chain structure of the zigzag interaction. In this paper, we thus call the 3D inter-chain coupling ‘3D coupling’.

susceptibility and the magnetization curve, and discuss generic features of them. In section 4 we show the experimental data of the magnetic susceptibility and the high-field magnetization parallel to the chain direction. Then the exchange coupling $J_2/k_B = 16.3$ K and the ratio $J_1/J_2 = 0.25$ are obtained by the precise comparison of the experimental data with the DMRG results. We also discuss the 3D effect and estimate the coupling $J' \sim 0.04J_2$. In section 5 we summarize our conclusions.

2. Computational and experimental details

2.1. Finite temperature DMRG

For the reliable analysis of the experimental results, systematic numerical data of the magnetization curve and the susceptibility are required in a wide parameter region. However, the quantitative calculation for the zigzag chain at finite temperatures has not been made yet, especially for the nearly two chain case. In this study we employ the finite temperature DMRG method [14, 15], which is free from the negative sign problem and thus enables us to calculate the quantities at sufficiently low temperatures with high accuracy. We note that the method has actually been applied successfully to some frustrated spin ladder systems [16, 18–20]. Following the implementation procedure described in [19], we calculate the magnetization and the susceptibility with the maximum number of retained bases $m = 240$. The lowest temperature we can reach is $k_B T/J = 0.07$, where J is the larger of the two exchange couplings J_1 and J_2 . We have confirmed that the computed data converge with respect to m and the Trotter number.

2.2. Experiment

We next summarize the synthesis and the crystal structure of $(\text{N}_2\text{H}_5)\text{CuCl}_3$. $(\text{N}_2\text{H}_5)\text{CuCl}_3$ samples were synthesized according to the method described in [12]. After several syntheses, we obtained small single crystal samples with a typical size of $1.5 \times 2 \times 3$ mm³. Chemical analysis shows good agreement between the observed and calculated ratios of the elements. This compound crystallizes in the orthorhombic system (space group: $Pnma$) [12]. The lattice constants at room temperature are $a = 14.439(2)$ Å, $b = 5.705(1)$ Å and $c = 6.859(1)$ Å. The chain structure of this compound is shown in figure 2. This compound has a ladder structure in which Cu and Cl align alternately along the leg, forming a copper zigzag chain. Copper 3d hole orbitals ($3d_{x^2-y^2}$) are situated perpendicularly to the leg, resulting in small exchange interactions along the leg.

Magnetic susceptibilities were measured with a SQUID magnetometer (Quantum Design's MPMS-XL7) installed at KYOKUGEN in Osaka University. As well as high-field magnetization measurements up to 30 T carried out with a pulse magnet, X-band ESR (Varian E109 spectrometer) and specific heat measurements were also performed at KYOKUGEN.

3. DMRG results

In this section, we present the DMRG results for the susceptibility and the magnetization curve, and then discuss the frustration dependence of such quantities for the nearly single chain and nearly decoupled chains cases.

Figure 3 shows the normalized susceptibilities $\chi^* \equiv \chi/\chi_{\max}$, where χ is the magnetic susceptibility per site and χ_{\max} is its maximum value. The normalized temperature $T^* \equiv$

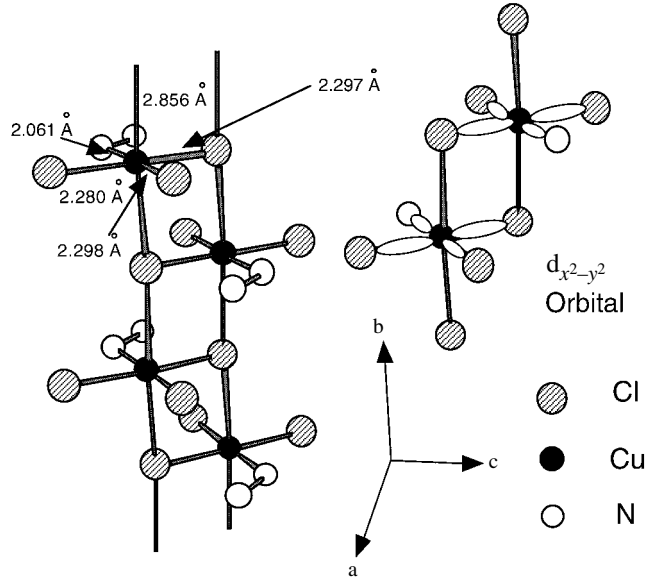


Figure 2. Chain structure of $(\text{N}_2\text{H}_5)\text{CuCl}_3$.

Table 1. The values of T_{\max} and χ_{\max} extracted from the DMRG results with the normalization $J_1 = 1$ for $J_1 > J_2$ and $J_2 = 1$ for $J_1 < J_2$. We also set $k_B = g\mu_B = 1$ for simplicity.

J_2/J_1	$J_1 > J_2$		$J_1 < J_2$		
	T_{\max}	χ_{\max}	J_1/J_2	T_{\max}	χ_{\max}
0.0	0.640	0.147	0.0	0.640	0.147
0.1	0.588	0.147	0.1	0.640	0.143
0.2	0.516	0.148	0.2	0.635	0.139
0.3	0.419	0.149	0.3	0.630	0.135
0.4	0.299	0.152	0.4	0.620	0.131
0.5	0.279	0.153	0.5	0.606	0.128

T/T_{\max} is also introduced, where T_{\max} is defined as the temperature at which χ indicates χ_{\max} . The values of T_{\max} and χ_{\max} are summarized in table 1.

Let us first consider the nearly single chain case ($J_1 > J_2$). In figure 3(a), we can see the feature of the gapless Heisenberg chain for $J_2/J_1 \leq 0.3$; in a high temperature region χ^* increases with decreasing T^* , and takes a round peak at $T^* = 1$. Then χ^* starts to decrease, and finally converges to a finite value as $T^* \rightarrow 0$. Although the spin fluid-dimer transition occurs at $J_2/J_1 = 0.2411$, the expected dimer gap is exponentially small for $J_2/J_1 = 0.3$ [8], where the almost gapless behaviour of χ^* appears [19]. As J_2/J_1 is increased to 0.4 and 0.5, the dimer-gapped phase starts to develop rapidly, and then the exponential decay of χ^* appears explicitly in the low temperature region. This rapid growth of the dimer gap also causes the considerable enhancement of χ^* in the high temperature side ($T^* > 1$) for $J_2/J_1 = 0.4$ and 0.5.

We next consider the nearly decoupled chains case ($J_1 < J_2$), where we can also see that χ^* basically shows the feature of the gapless chain. A remarkable point, in contrast to the nearly single chain case, is that the gapless-like behaviour is observed in a wide range of J_1/J_2 (see figure 3(b)) and thus the dependence of χ^* on J_1/J_2 is relatively weak. This wide-range gapless-like behaviour is attributed to the extremely small spin gap,

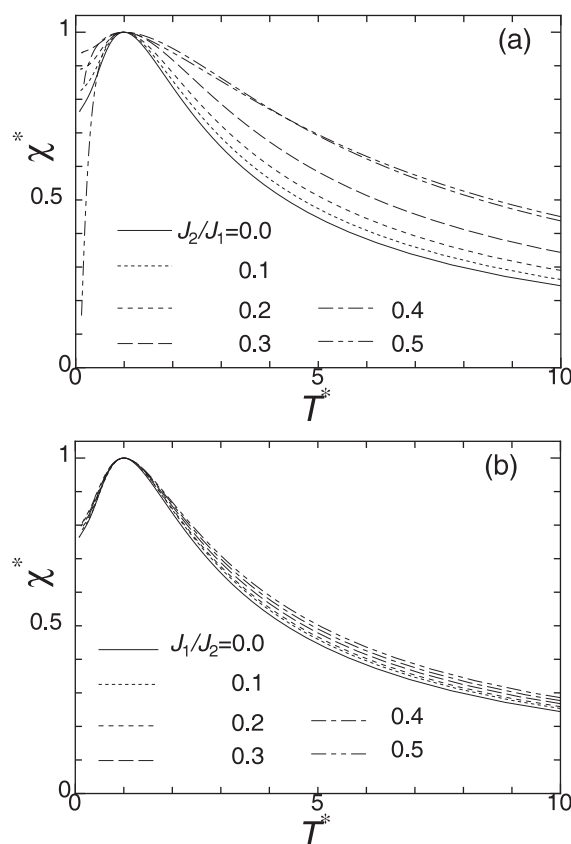


Figure 3. Finite temperature DMRG results of the susceptibility (a) for $J_1 > J_2$ and (b) for $J_1 < J_2$.

$\Delta \sim \exp(-\text{constant} \times J_2/J_1)$, although the system is considered to be the dimer-gapped phase for $J_1 \neq 0$. For example, the spin gap is estimated as $0.03J_2$ even at $J_1/J_2 = 0.5$ [9], and thus the effect of the gap is easily smeared out by the finite temperature effect.

Here, we wish to point out an important feature of the susceptibility when analysing the experimental results. If the frustration is small, it may be difficult to discriminate between the behaviour of the nearly single chain and that of the nearly decoupled chains only from the susceptibility result, because χ^* of the nearly single chain and that of the nearly decoupled chains are very similar to each other. However, an important point is that the frustration dependence of T_{max} for $J_1 > J_2$ is relatively sensitive to J_2/J_1 (see table 1), implying that the energy scale of the dominant coupling constants is different between $J_1 > J_2$ and $J_1 < J_2$, even if the shapes of the susceptibility are quite similar to each other. For example, the shape of χ^* for $J_2/J_1 = 0.1$ is almost coincident with that for $J_1/J_2 = 0.3$, but the values of T_{max} for $J_2/J_1 = 0.1$ and $J_1/J_2 = 0.3$ are 0.588 and 0.630 respectively, which show about 7% deviation.

We now proceed to the results of the magnetization curves. Figure 4 shows the magnetization curves both for $J_1 > J_2$ and for $J_1 < J_2$ at $k_B T/J_1$ (J_2) = 0.125, which can be directly compared with experimental results of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ measured at 2 K as shown later. In the figure, we can basically see the characteristic property in the gapless Heisenberg chain as well; after the linear behaviour with H is observed near the zero magnetic

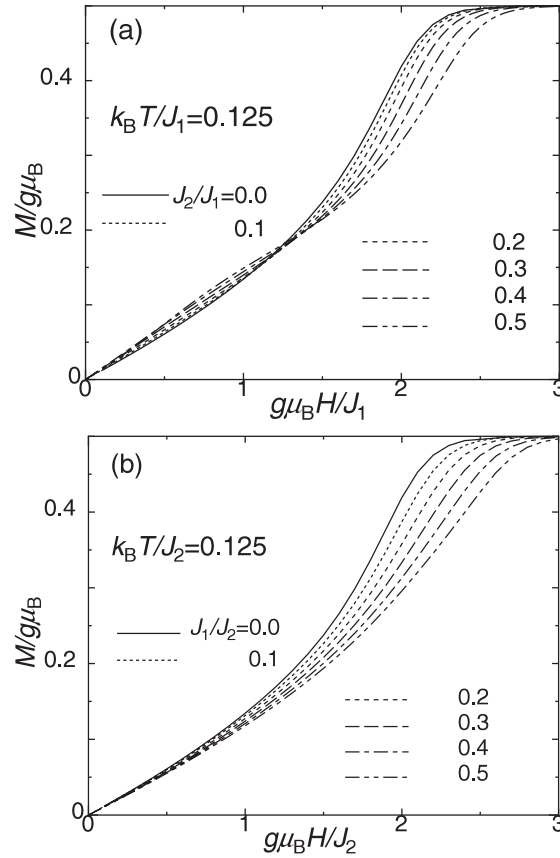


Figure 4. The magnetization curves obtained with the DMRG (a) for $J_1 > J_2$ at $k_B T/J_1 = 0.125$ and (b) for $J_1 < J_2$ at $k_B T/J_2 = 0.125$.

field ($H = 0$), the magnetization M increases rapidly in the high-field region, and finally saturates near $g\mu_B H/J_1(J_2) \sim 2.5$. However, what we want to remark here is that quantitative differences appear in the magnetization curve. For instance, in figure 4(a), we can see that the growth of the dimer gap for $J_2/J_1 \geq 0.4$ causes the enhancement of the magnetization in the middle-field region ($g\mu_B H/J_1 \simeq 1$), which is consistent with the susceptibility result. Moreover the magnetization curve around the saturation field H_s is shifting to the high-field side systematically as the frustration increases.

In order to capture the feature of the magnetization curves in each region of $J_1 > J_2$ and $J_1 < J_2$, we focus here on the above mentioned systematic shift of the magnetization curve near the saturation field H_s . For the zigzag chain, the behaviour of the saturation field H_s is easily obtained as

$$H_s = \begin{cases} \frac{2J_1}{g\mu_B} & \text{for } J_2/J_1 < 1/4 \\ \frac{1}{g\mu_B} \left(J_1 + 2J_2 + \frac{J_1^2}{8J_2} \right) & \text{for } J_2/J_1 > 1/4. \end{cases} \quad (2)$$

For the nearly single chain, the saturation field H_s stays at $2J_1/g\mu_B$ for $J_2/J_1 < 1/4$, and it starts to increase gradually as J_2/J_1 increases beyond $1/4$. On the other hand, for the

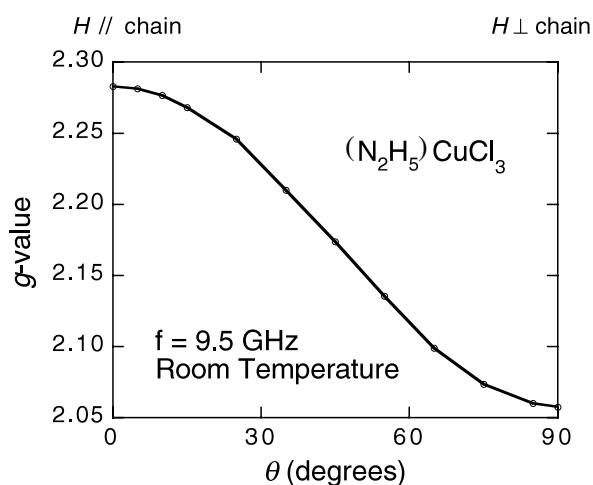


Figure 5. The angular dependence of the g -value of $(\text{N}_2\text{H}_5)\text{CuCl}_3$.

nearly decoupled chains, H_s starts to increase linearly as soon as the inter-chain coupling is introduced. This dependence of H_s readily appears in the magnetization curve shown in figure 4; the magnetization curves near H_s for $J_2/J_1 < 0.3$ remain in the ‘vicinity’ of the pure Heisenberg one. In contrast, for the nearly double chain, we can see that the curves near H_s shift to the high-field side linearly with respect to J_1/J_2 . As a result, the magnetization curves near H_s for $J_1 > J_2$ become slightly steeper than those for $J_1 < J_2$.

As was seen for the susceptibility result, the dominant coupling determined from the susceptibility exhibits a slightly different value depending on the nearly single chain or nearly decoupled chains cases. Then such difference of the dominant coupling affects the magnetization curve near the saturation field, which can be resolvable with the comparison between the accurate DMRG result and the experimental ones. Therefore the coupling constant of the zigzag chain can be expected to be determined with the cooperative use of the susceptibility and the magnetization curve.

4. Experimental results

4.1. ESR and specific heat

We performed X-band ESR measurements at room temperature on a single crystal sample of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ in order to get the g -values precisely. Accurate comparison between the experimental data and the numerically calculated ones requires precise g -values. Figure 5 shows the angular dependence of the g -value. We obtained g -values of 2.285 and 2.060 for the external magnetic field parallel and perpendicular to the chain, respectively. In the following comparisons between experiments and calculations, we used these g -values.

We carried out specific heat measurements on a powder sample of $(\text{N}_2\text{H}_5)\text{CuCl}_3$. Figure 6 shows the temperature dependence of the specific heat divided by temperature. Here, the data include the lattice contribution. We observed an anomaly at 1.55 K (T_N) which is probably due to the AF long-range ordering. In order to avoid the 3D effect in this quasi-one-dimensional system, high-field magnetization experiments were done at 2 K above T_N .

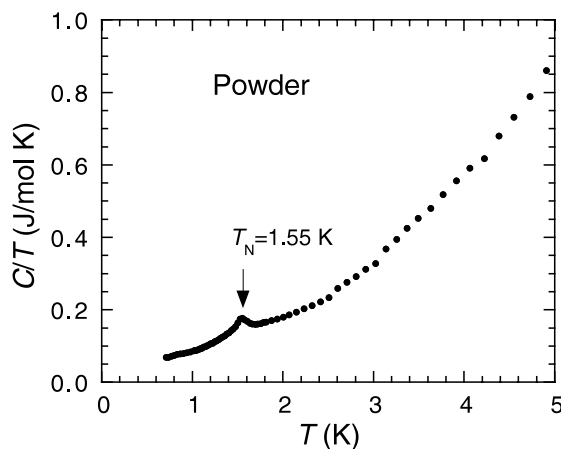


Figure 6. The specific heat of a powder sample of $(\text{N}_2\text{H}_5)\text{CuCl}_3$.

4.2. Magnetic susceptibility

Magnetic susceptibility χ_{EXP} of the single crystal of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ parallel to the chain, which was used for the ESR measurement, is presented in figure 7, where the DMRG results χ_{DMRG} are also shown for comparison. In the high temperature region the χ_{EXP} exhibits the Curie–Weiss-type T -dependence, and has a round peak around $T = 10$ K. When the temperature is decreased further, χ_{EXP} also decreases and finally converges to a finite value in the zero temperature limit. Since these features are characteristic in gapless 1D antiferromagnets, we should take account of the two possibilities of gapless zigzag chains, as mentioned in the introduction. We thus analyse χ_{EXP} with the DMRG results both for the single chain limit ($J_1 > J_2$) and the weakly coupled Heisenberg chain limit ($J_1 < J_2$). We first estimate the dominant exchange coupling constant from the position of the broad peak, by using $g = 2.285$ obtained from the ESR, and then tune another exchange coupling constant so as to reproduce the shape of the susceptibility well.

For the former case ($J_1 > J_2$), we find that χ_{DMRG} of $J_2/J_1 = 0.1$ with $J_1/k_B = 17.4$ K agrees well with χ_{EXP} in the whole temperature region (see figure 7(a)). For the latter ($J_1 < J_2$), we can also see that the DMRG results for $0.2 < J_1/J_2 < 0.3$ well reproduce the shape of the experimental susceptibility, as in figure 7(b), where the best fitted value is obtained as $J_1/J_2 = 0.25$ with $J_2/k_B = 16.3$ K. Since the DMRG results seem to explain the whole shape of the susceptibility well for both cases, it is unfortunately difficult to confirm which case is realized in $(\text{N}_2\text{H}_5)\text{CuCl}_3$ only from the susceptibility result, as discussed in the previous section. However, we note that the dominant coupling $J_1/k_B = 17.4$ K for $J_1 > J_2$ is about 6% bigger than $J_2/k_B = 16.3$ K for $J_1 < J_2$, which is consistent with the DMRG result. For the case of the $H \perp$ chain, very similar results were obtained using the same parameter values of the exchange couplings as for the $H \parallel$ chain and $g = 2.060$ from ESR.

Before proceeding to the next subsection, we make a comment about a possibility of the weak ferromagnetic coupling. We have also computed the case of a negative $J_2(J_1)$ for $J_1 > J_2$ ($J_1 < J_2$). However, the shape of the susceptibility is changed drastically by introducing the ferromagnetic coupling, so that the couplings of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ are AF.

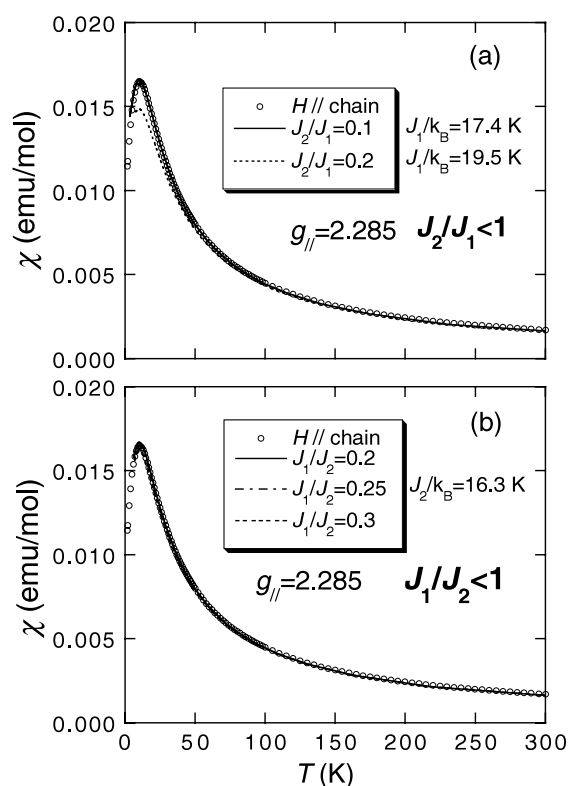


Figure 7. Magnetic susceptibility of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ compared with the DMRG results (a) for $J_1 > J_2$ and (b) for $J_1 < J_2$. The open circles denote experimental data χ_{EXP} and the curves show finite temperature DMRG results χ_{DMRG} .

4.3. Magnetization curve

Figure 8 shows the magnetization curve for the single crystal of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ along the chain at $T = 2.0$ K up to 30 T. The temperature was set a little above T_N (1.55 K) as already mentioned. Around the saturation field, there is a small hysteresis of magnetization between the ascending and descending processes, due to the magnetocaloric effect. In the figure, we can see that the measured curve of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ is qualitatively similar to the gapless chain, which is consistent with the susceptibility results. Using the parameters estimated in the previous subsection, we now discuss the magnetization processes of the DMRG and the experiments. The temperature used in the numerical calculations was determined close to that in the experiments.

The results of comparisons are also shown in figure 8, where the DMRG curves both for $J_1 > J_2$ and for $J_1 < J_2$ are in good agreement with the experimental one in the low-field region. However, the magnetization curve in the high-field region is reproduced well by the coupling of $J_1/J_2 = 0.25$, as can be seen in figure 8(b). Indeed the gradient of the calculated curve for $J_1 > J_2$ is slightly bigger than the experimental one around $15 < H < 25$ T, while the curve for $J_1 < J_2$ is well fitted for the whole range of the magnetic field. The difference between two cases described in the previous section certainly appears in high-field region. Hence we have determined that the zigzag chain compound $(\text{N}_2\text{H}_5)\text{CuCl}_3$ is a weakly coupled double chain system with the parameters $J_1/J_2 = 0.25$ and $J_2/k_B = 16.3$ K.

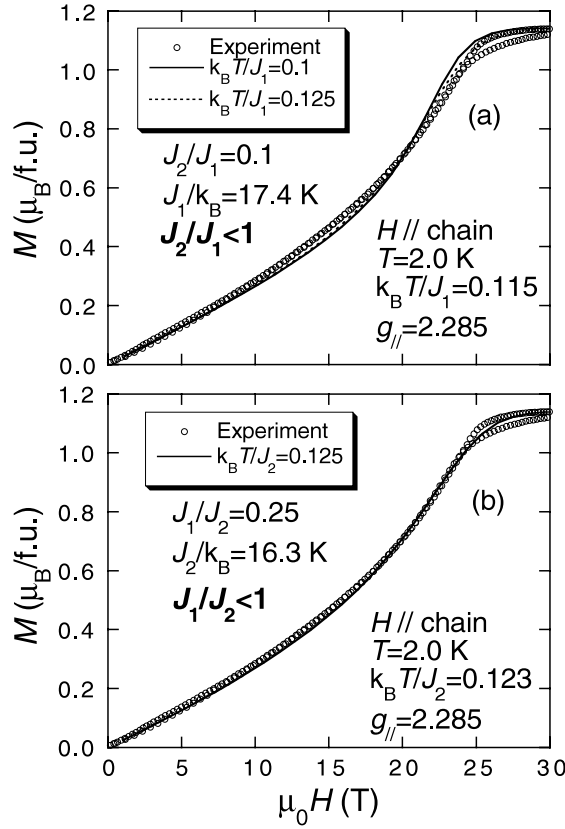


Figure 8. The magnetization curves of $(\text{N}_2\text{H}_5)\text{CuCl}_3$. The comparison with the DMRG results (a) for $J_2/J_1 = 0.1$ and (b) for $J_1/J_2 = 0.25$.

4.4. 3D interaction

In this subsection, we would like to discuss the effect of the 3D interaction on $(\text{N}_2\text{H}_5)\text{CuCl}_3$. Since the specific heat shows the 3D ordering at $T_N = 1.55$ K, some consideration for the 3D coupling may be required. To this end, we employ here the mean-field theory combined with the DMRG method [16, 17]. We use the obtained values of the couplings $J_1/J_2 = 0.25$ and $J_2/k_B = 16.3$ K, and assume a 3D Heisenberg-type interaction whose coupling constant is denoted as J' . According to the strong leg interaction J_2 , we assume a staggered magnetic field along the leg direction, and then determine the assumed magnetic field and the DMRG-calculated magnetic order self-consistently. In particular, the phase transition point within the mean-field theory is given by

$$\chi_s(T_N) = \frac{1}{zJ'} \quad (3)$$

where $\chi_s(T)$ is the staggered susceptibility and z is the coordination number. Calculating χ_s with the finite temperature DMRG method, we can estimate J' from the experimentally determined Néel temperature T_N through the equation (3). In figure 9, we plot the normalized staggered susceptibility $\chi_s^* \equiv J_2\chi_s$ obtained with finite temperature DMRG. In the figure, we find that the crossing point of χ_s^* and the experimentally obtained Néel temperature is given by $\chi_s^* = 6.6$ and $T_N^* \equiv k_B T_N/J_2 \sim 0.095$, where T_N^* is the normalized transition

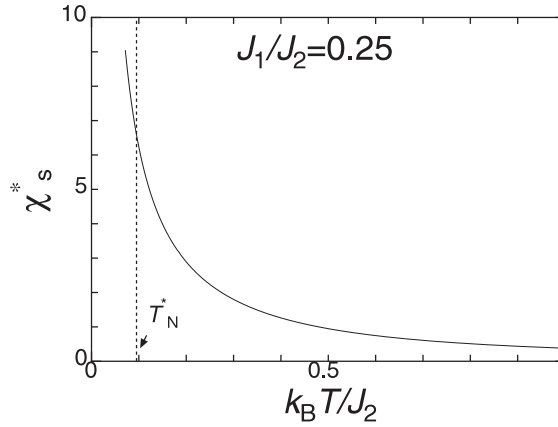


Figure 9. Staggered susceptibility of the zigzag chain for $J_1/J_2 = 0.25$. The dotted line denotes the normalized transition temperature $T_N^* \equiv k_B T_N/J_2 \sim 0.095$ for $(\text{N}_2\text{H}_5)\text{CuCl}_3$. The value of χ_s^* at $k_B T/J_2 = T_N^*$ gives the coupling zJ' following equation (3).

temperature. Assuming $z = 4$, we obtain $J' \sim 0.04J_2$ (~ 0.6 K), which is about $0.2J_1$. Thus we can conclude that the 3D interaction effect is much smaller than the intra-chain couplings. Therefore the zigzag chain picture is valid for $(\text{N}_2\text{H}_5)\text{CuCl}_3$ above the Néel temperature.

5. Summary and discussion

We have investigated the magnetic properties of $(\text{N}_2\text{H}_5)\text{CuCl}_3$, which can be regarded as a zigzag chain with the coupling $J_1 < J_2$, by the accurate comparison of the numerical calculations and experiment results.

First, we have calculated the susceptibility and the magnetization curves of the zigzag chain using the finite temperature DMRG method for the nearly single chain case $J_1 > J_2$ and for the nearly double chain case $J_1 < J_2$ to clarify the differences between the two cases appearing in the parameter dependence of the physical quantities. From the experimental point of view, the quantitative difference appearing in the high-field region of the magnetization curve is found to help us to identify the magnetic nature of $(\text{N}_2\text{H}_5)\text{CuCl}_3$.

We have next presented the experimental results for a single crystal of $(\text{N}_2\text{H}_5)\text{CuCl}_3$. The g -values parallel and perpendicular to the chain are determined by the ESR experiment as $g = 2.285$ and 2.060 , respectively. The Néel temperature is also estimated as $T_N = 1.55$ K from the specific heat of a powder sample. Above the Néel temperature, the observed susceptibility and the magnetization for the single crystal show gapless or nearly gapless behaviour. An accurate comparison of the experimental data with the DMRG results shows that the DMRG results for $J_1/J_2 = 0.25$ with $J_2/k_B = 16.3$ K reproduce the experimental results well. The estimated couplings also show that the dimer gap of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ is $0.001J_2 \sim 0.02$ K [9], which is much smaller than T_N . Hence the gapless behaviour of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ observed above T_N is consistent with the obtained exchange couplings.

Finally, we have discussed the effect of the 3D interaction, using the mean-field theory combined with the finite temperature DMRG method. The estimated value of the 3D coupling J' ($\sim 0.04J_2$) is sufficiently small compared with the intra-chain couplings, which supports the zigzag chain picture of the compound.

In the present paper, we have addressed the zigzag compound of $(\text{N}_2\text{H}_5)\text{CuCl}_3$ in terms of the bulk quantities: the magnetic susceptibility and the high-field magnetization, for

which rather precise analyses are required. However, the frustration effect often gives rise to incommensurate behaviour in the correlation function [9, 21], which is difficult to capture in the bulk quantities. Thus it is also an interesting problem to investigate the compound by measurements of the fluctuation-associated quantities with position resolvable methods, such as NMR or the inelastic neutron scattering.

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