# Magnetic Properties of Bond-Alternating Quantum Spin Chain System: $(CH_3)_2NH_2CuX_3$ (X=Cl,Br)

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**Abstract**

Magnetic properties of ferromagnetic(F) and antiferromagnetic(AF) bond alternating quantum spin chain system, $DMACuX_3$ where X=Cl, Br and DMA=$(CH_3)_2NH_2$, are reported. The most prominent result is that for Cl-compound we observed the unusual mixed state in which $S$=1 and $S$=0 dimers coexist evenly due to frustrated situation in the presence of comparable F and AF couplings. For Br-compound, it is shown from the susceptibility measurements that an AF coupling is dominant and it provides an example of simple dimer-limit.

*Key words:* Quantum spin chain; Bond alternation; $(CH_3)_2NH_2CuCl_3$; $(CH_3)_2NH_2CuBr_3$

The magnetic behavior of uniform spin 1/2 quantum Heisenberg chains has been well documented, for both ferromagnetic(F) and antiferromagnetic(AF) exchange couplings. With the presence of dimerization of the chain, we expect interesting systems with alternating nearest exchange couplings, $J_1$ and $J_2$. Of particular interest is the system with a negative alternating parameter $\alpha$=$J_2/J_1$ in which F/AF bond alternation within a chain is realized. In a limit of dominant F coupling, $S$=1 dimer is formed and the system behaves like a Haldane system of $S$=1 AF chain at low temperatures, having a characteristic nonmagnetic singlet-ground-state (SGS) with a spin gap (Haldane-limit). On the other hand, in an opposite limit of dominant AF coupling, $S$=0 dimer is formed and the system behaves like a weakly F-coupled $S$=1/2 AF dimerized system, having a trivial nonmagnetic SGS with a spin gap (Dimer-limit). Hida has shown that two limits are continuously connected to each other without any singularity; they are in the same phase, at least, in the ground state [1].

Even though, here, we are interested in an intermediate regime between the Haldane-limit and Dimer-limit. Our simple motivation is to inquire what happens in a frustrated situation, in some sense, due to the presence of comparable F and AF couplings. Our model substances are $DMACuX_3$ where DMA=$(CH_3)_2NH_2$ and X=Cl, Br, in which stractural pathways show that $Cu^{2+}$ dimers form chains, with each $Cu^{2+}$ ion bridging to a halide ion in an adjacent dimer, yielding an alternating chain [2]. There are several experiments of this compound [3–6] but its magnetic properties are still controversial.

(1) *$DMACuCl_3$*

Figs. 1 and 2 show the temperature dependencies of magnetic susceptibility and specific heat, respectively. The measured susceptibility monotonically increases with decreasing temperature. At a first glance, it looks a usual paramagnetic behavior showing a Curie-like temperature dependence. However, it is not the case as evidenced by $\chi T$-$T$ plot, showing a gradual rise in

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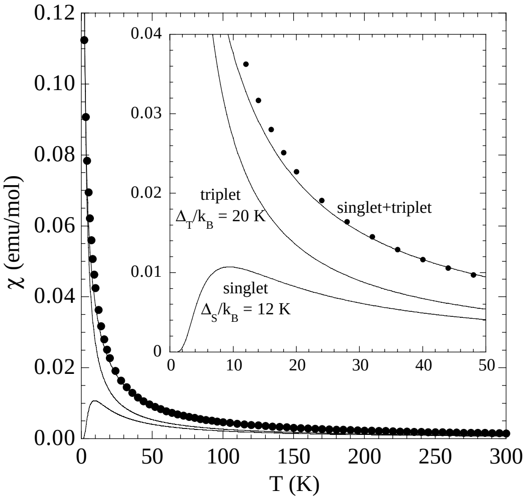


Fig. 1. Tempereture dependence of magnetic susceptibility. Calculated results are shown by the solid lines, together with the each contribution from the singlet and triplet units.

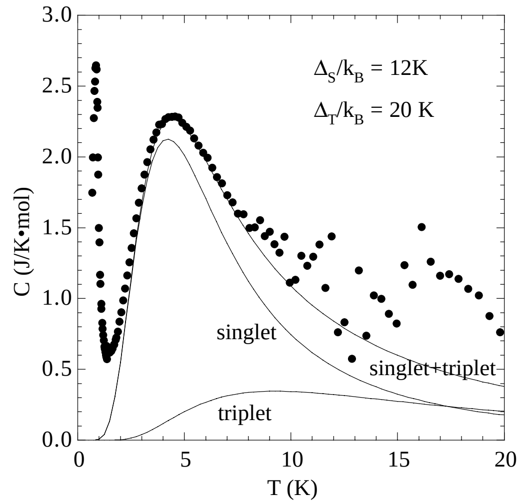


Fig. 2. Tempereture dependence of magnetic specific heat. Calculated results are shown by the solid lines, together with the each contribution from the singlet and triplet units.

$\chi T$ as the temperature initially decreases and a precipitous drop at low temperature, characteristic of a magnetic system with a dominant F coupling and a weaker AF coupling. The magnetic specific heat exhibits a broad maximum around 4 K characteristic of one-dimensional (1D) system, besides a sharp peak at 0.8 K, originating from the 3D magnetic transition.

Numerical calculation of the thermodynamic properties of an alternating F/AF Heisenberg chain is now available up to ring chain of 14 spins [7]. Unfortunately, however, we did not find any consistent sets of F and AF couplings which yield a reasonable fit to the both data at the same time. We think that this difficulty might be an indication of some frustration effect. Therefore, we introduce a simplified model to explain the essence of our results; that is a naive model in terms of quasi-noninteracting two subunits. One subunit forms an AF coupled dimer having a $S$=0 singlet ground state and an excited $S$=1 triplet state with an energy gap $\Delta_{\mathrm{S}}$, and, conversely, the other forms a F coupled dimer having a $S$=1 ground state and an excited $S$=0 state with an energy gap $\Delta_{\mathrm{T}}$. As shown by the solid lines in Figs. 1 and 2, we obtain an excellent fit to the both data simultaneously by simply additing two contributions from subunits, using $\Delta_{\mathrm{S}}/k_{\mathrm{B}}$=12 K and $\Delta_{\mathrm{T}}/k_{\mathrm{B}}$=20 K with an equal probability for two subunits.

If the above scenario is the case, it may be concluded the existence of the unusual mixed state, in which $S$=1 and $S$=0 dimers coexist at an equal probability, probably dynamically, in a frustrated situation due to the presence of comparable F and AF couplings.

High-field magnetization under pulse field up to 30 T down to 0.5 K is also peculiar. At the lowest temperature it increases rapidly to a 1/2 plateau at 2 T and after keeping a constant value up to 3.5 T it increases gradually with up-turn curvature to the saturation at 14 T. Detail discussion is reported elsewhere.

(2) *DMACuBr*$_3$

Magnetic susceptibility shows a broad maximum around 70 K. It was concluded that an AF coupling is dominant with AF coupling of 56 K and alternation ratio of 0.2. Br-compound provides an example of Dimer limit of alternating chain.

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