IPA-CuCl$_3$: a $S = 1/2$ Ladder with Ferromagnetic Rungs

T. Masuda[1*], A. Zheludev[*], H. Manaka[†] and J.-H. Chung[**]

[*]*Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6393, USA.*
[†]*Department of Nano-Structure and Adv. Materials, Kagoshima University, Kagoshima 890-0065, Japan.*
[**]*NCNR, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA.*

Abstract. The spin gap material IPA-CuCl$_3$ has been extensively studied as a ferromagnetic-antiferromagnetic bond-alternating $S = 1/2$ chain. This description of the system was derived from structural considerations and bulk measurements. New inelastic neutron scattering experiments reveal a totally different picture: IPA-CuCl$_3$ consists of weakly coupled spin ladders with antiferromagnetic legs and ferromagnetic rungs. The ladders run perpendicular to the originally supposed bond-alternating chain direction. The ferromagnetic rungs make this system equivalent to a Haldane $S = 1$ antiferromagnet. With a gap energy of 1.17(1) meV, a zone-boundary energy of 4.1(1) meV, and almost no magnetic anisotropy, IPA-CuCl$_3$ may the best Haldane-gap material yet, in terms of suitability for neutron scattering studies in high magnetic fields.

Despite years of extensive research, a number of very important models of one-dimensional (1D) magnets have not yet found their realizations in real quasi-1D materials. This "deficit" became particularly frustrating when recent attention turned to neutron studies of high-field effects in quantum magnets [1, 2, 3, 4, 5]. Most known spin-ladder compounds have very large scales of magnetic interactions and are unsuitable for such studies. A number of Haldane-gap and related systems with small enough gap energies have been investigated [1, 2, 6], but all feature very strong single-ion anisotropy that qualitatively alters high-field behavior. For this reason, the discovery of the quasi-1D material (CH$_3$)$_2$CHNH$_3$CuCl$_3$ (IPA-CuCl$_3$) with almost no anisotropy and a spin gap of ~ 1.5 meV was a major step forward.[7, 8, 9] Bulk measurements on this material revealed a field-induced Bose condensation of magnons at a critical field $H_c \simeq 9$ T [8]. The original indication was that IPA-CuCl$_3$ should be described in terms of $S = 1/2$ Cu^{2+} -chains running along the c axis of the triclinic structure (dashed line in Fig. 1). Moreover, it appeared that the exchange interactions connecting subsequent Cu^{2+} spins alternate between ferromagnetic and antiferromagnetic ones. Herein we report neutron scattering studies of the magnetic excitations in IPA-CuCl$_3$ that reveal a totally different picture.

Fully deuterated IPA-CuCl$_3$ samples were utilized for these studies. 20 single crystals of total mass 3.5 g were co-aligned to a cumulative mosaic of 1.5°. Magnetic susceptibility and ESR data are similar to those for the
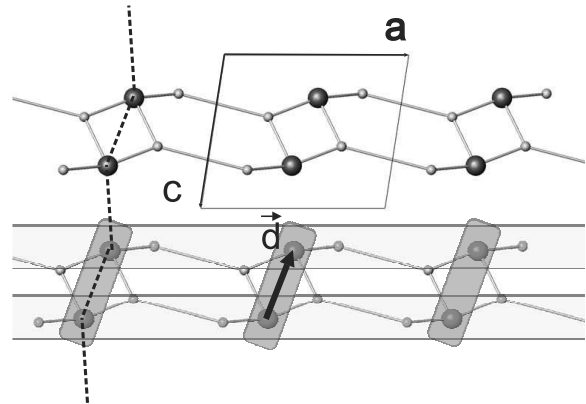

FIGURE 1. Schematic view of the spin-ladder structure of IPA-CuCl$_3$ in projection onto the (a, c) crystallographic plane. Large balls are magnetic Cu^{2+} ions. Small balls are Cl$^-$. The previously proposed alternating-chain model, now shown to be incorrect, is represented by a dashed line.

non-deuterated material. Cell parameters at room temperature are $a = 7.766$ Å, $b = 9.705$ Å, $c = 6.083$ Å, $\alpha = 97.62°, \beta = 101.05°$ and $\gamma = 67.28°$, space group $P - 1$ [7]. The measurements were done on the SPINS cold neutron 3-axis spectrometer at NIST, using $E_f = 3.7$ meV fixed-final-energy neutrons and a BeO filter positioned after the sample. The scattering plane was either $(h, 0, l)$ or $(h, k, 0)$. Most data were taken at $T = 1.5$ K.

From the very first scans it became obvious that c^* is *not* the direction of strong dispersion. This implies that the bond-alternating c-axis chain scenario is *not* an appropriate model for IPA-CuCl$_3$, and that the spin gap

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[1] Present address: Department of Basic Science, Yokohama City University, Japan.
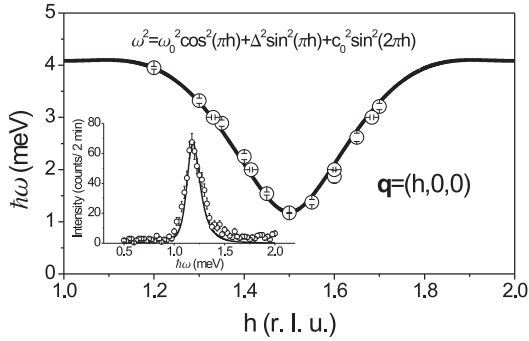
FIGURE 2. Measured a-axis dispersion of the ladder gap excitations in IPA-CuCl$_3$. The measured bandwidth along the c crystallographic axis is only about 0.6 meV [11]. Inset: constant-q scan collected at the 1D antiferromagnetic zone-center $(0.5, 0, 0)$.

must have a different origin. A systematic survey of the spectrum enabled us to locate the global energy minimum for the gap excitations at $\mathbf{q} = (0.5, 0, 0)$. A typical energy scan at this wave vector is shown in the inset of Fig. 2. The observed peak position is in good agreement with the gap energy deduced from bulk measurements.

Additional measurements established that IPA-CuCl$_3$ is indeed one-dimensional magnet, but with the a-axis being the strong-coupling direction. The dispersion relation of the gap excitations along a^* was measured in a series of const-q and const-E scans. The resulting dispersion curve is plotted in Fig. 2. These data were analyzed using the empirical formula shown in the figure[10]. A good fit was obtained using $\hbar\omega_0 = 4.1(1)$ meV $\Delta = 1.17(1)$ meV and $c_0 = 2.2(1)$ meV (solid line). The energy of the observed gap mode was also measured as a function of momentum transfer along the other two reciprocal-space directions. The bandwidth along c^* is rather narrow, the gap increasing from its minimum $\Delta = 1.17$ meV at $\mathbf{q} = (0.5, 0, 0)$ to a maximum value of 1.8 meV at $\mathbf{q} = (0.5, 0, 0.5)$. There is no measurable k-dependence of the gap energy. More details will be reported elsewhere [11].

The positions of the magnetic Cu^{2+} ions in IPA-CuCl$_3$ are non-alternating along the crystallographic a axis. A careful look at the structure allows to identify uniform *spin ladders* running along this direction, as shown in Fig. 1. The structural alternation along the c axis that was the basis of the alternating-chain model ensures a separation between individual ladders. Along b the ladders are even better isolated due to non-magnetic layers of the organic ligand. The ladder model was confirmed by studying the wave vector dependence of integrated intensities of the gap modes. At the 1D AF zone-centers $h = 0.5$ and

$h = 1.5$ this intensity is periodic with l and scales precisely as $\cos^2(\mathbf{qd}/2)$, where $\mathbf{d}$ is the vector that defines the ladder rung (Fig. 1) [11]. Such a 3D structure factor implies that the rung interactions are *ferromagnetic*, as previously proposed [7]. A ladder of this type can be viewed as a Haldane spin chain, each pair of rung spins playing the role of an effective $S = 1$. This is especially true for excitations at wave vectors with $\mathbf{qd} = 0$, where the dynamic structure factor is exactly equal the correlation function for the total spin on each rung. We conclude that IPA-CuCl$_3$ can be described as quasi-1D antiferromagnet with "composite" Haldane spin chains along a.

IPA-CuCl$_3$ turned out not to implement the bond-alternating chain model as it was assumed to do for quite some time. Nevertheless, it remains a very interesting and important low-dimensional material. Future high-field neutron scattering studies of this anisotropy-free composite Haldane spin chain system will be interesting to contrast with recent work on the strongly anisotropic Haldane gap material NDMAP [2] and the bond-alternating $S = 1$ chain compound NTENP [6].

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