Magnetization of staggered $S = 1/2$ antiferromagnetic Heisenberg chain systems

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Abstract. Cu Pyrimidine Cu(C$_4$N$_2$H$_4$)(NO$_3$)$_2$(H$_2$O)$_2$ and Cu Benzoate Cu(C$_6$H$_5$COO)$_2$·3H$_2$O are molecule-based $S = 1/2$ antiferromagnetic Heisenberg chain systems. Because of a staggering of the $g$ tensor and the Dzyaloshinskii-Moriya interaction, the magnetic principal axis system $a''bc''$ is distinct from the crystallographic unit cell. Along $a''$ a behavior corresponding to the uniform $S = 1/2$ antiferromagnetic Heisenberg chain occurs, while along $c''$ the effect of staggering of the $g$ tensor and Dzyaloshinskii–Moriya interaction are most prominent. Here, we discuss the field and temperature dependent magnetization along $a''$ and $c''$ of these systems, in particular with respect to the relevance of the inhomogeneity parameter $k$ controlling the ratio between longitudinal and transverse magnetization components.

Because of a rich variety of different magnetic ground states such as quantum critical behavior or gaps in the spin excitation spectra, one–dimensional quantum magnets have been of much interest to theorists and experimentalists alike [1-5]. In recent years, in particular systems lacking inversion symmetry in the crystallographic lattice have been studied in detail. Because of the low symmetry, besides of a Heisenberg exchange additional terms, viz., an alternation of the $g$ tensor and the Dzyaloshinskii–Moriya (DM) interaction, may have to be taken into account in order to describe such materials (exchange anisotropies are neglected in this approach) [6,7]. In consequence, for materials like Cu Pyrimidine Cu(C$_4$N$_2$H$_4$)(NO$_3$)$_2$(H$_2$O)$_2$ and Cu Benzoate Cu(C$_6$H$_5$COO)$_2$·3H$_2$O [8-11] the effective Hamiltonian is written as

$$H = J \sum_i \vec{S}_i \vec{S}_{i+1} - h_u S_i^z - (-1)^i h_s S_i^z,$$

(1)

with the coupling constant $J$, the effective uniform field $h_u = g \mu_B H / J$ and the induced effective staggered field $h_s = k h_u$. For a given field axis the effective $g$ and the inhomogeneity parameter $k$ are determined from the alternating $g$ tensor $\vec{g} = \vec{g}_u \pm \vec{g}_s$ and the DM interaction $\vec{D}$

$$g = \left| \vec{g}_u \vec{H} \right| / \left| \vec{H} \right|,$$

(2)

$$k = \frac{1}{g} \left| \frac{1}{2J} \vec{D} \times \vec{g}_u \vec{H} + \vec{g}_s \vec{H} \right| / \left| \vec{H} \right|.$$

(3)
In essence, for these materials the presence of DM interaction and staggered $g$ tensor induces new symmetry breakings with respect to the magnetic properties \[6,7\]. In terms of principal axes, it is distinguished between the crystallographic unit cell with the axes $abc$ and the system of the principal magnetic axes $a''b''c''$ (for the procedure to derive the coordinate frame $a''b''c''$ from the local crystallographic symmetry in these low dimensional materials see Refs. \[7,8\]). Now, a magnetic behavior along the direction of $a''$ corresponding to that of the uniform spin $S = 1/2$ antiferromagnetic Heisenberg chain ($S = 1/2$ AFHC) is observed \[12,13\], while the effects of staggering of the $g$ tensor and DM interaction are most prominent along $c''$. For both Cu Pyrimidine and Cu Benzoate the $g$ tensor and the DM vector have been derived previously, as have the different coordinate frames. Correspondingly, these quantum spin systems allow a quantitatively exact comparison between theory and experiment. In particular, this has been demonstrated for the magnetization, where both the temperature and field dependence have been determined experimentally over a wide range in reduced temperatures $k_B T/J$ and effective fields $g \mu_B H/J$, and have been compared to calculations based on the Hamiltonian presented in Eq. (1) \[14,15\].

In this contribution, we combine the experimental findings on both compounds, Cu Pyrimidine and Cu Benzoate, by presenting the results of magnetization measurements for magnetic fields directed along $a''$ and $c''$ axes. The data are compared to calculations based on renormalization group analysis for transfer matrices (TMRG) and exact diagonalization (ED) of finite spin chains. From this analysis, we derive the field dependence of the induced staggered component of the magnetization. In particular, we discuss the dependence of the staggered magnetization on the inhomogeneity parameter $k$.

![Figure 1](image)

**Figure 1.** The reduced magnetization $M/M_{sat}$ of Cu Pyrimidine and Cu Benzoate as function of reduced magnetic field $\mu_0 H/\mu_0 H_{sat}$ along the magnetic principal axes $a''$ (a) and $c''$ (b,c) for reduced temperatures $k_B T/J = 0.04$ (Cu Pyrimidine) and 0.09 to 1.32 (Cu Benzoate). The solid lines represent the calculated magnetization at $T = 0$ for the uniform $S = 1/2$ AFHC (a) and the staggered model using an inhomogeneity parameter $k = 0.11$ (c); for details see text.

In Fig. 1 we plot the reduced magnetization $M/M_{sat}$ of Cu Pyrimidine and Cu Benzoate as function of reduced magnetic field $\mu_0 H/\mu_0 H_{sat}$, measured along the magnetic principal axes $a''$ and $c''$ for reduced temperatures $0.04 \leq k_B T/J \leq 1.32$. Details of the experiments are presented in the Refs. \[14,15\]. For Cu Pyrimidine the values for $M_{sat}$ are 1.07 and 1.11$\mu_B$/Cu atom for saturation fields $\mu_0 H_{sat}$ of 50.1 and 48.5 T along $a''$ and $c''$ axis, respectively.
Correspondingly, for Cu Benzoate the values are \( M_{\text{sat}} = 1.06(1.16) \mu_B/\text{Cu atom} \) and \( \mu_0 H_{\text{sat}} = 26.5(24.2) \) T along \( a'' \) \( (c'') \) axis [15]. The magnetic coupling strength \( J/k_B \) of Cu Pyrimidine is reported to 36.5 K, while for Cu Benzoate we have a value of 19 K. For comparison, we include the calculated magnetization at \( T = 0 \), as obtained for the direction \( a'' \) via Bethe ansatz [16] and along \( c'' \) from exact diagonalization of linear chains with \( N = 20 \) spins, using here an anisotropy parameter \( k = 0.11 \) previously determined for Cu Pyrimidine [14].

The plot highlights the qualitative difference of the field dependence of the magnetization at low temperatures \( k_B T \ll J \) between measurements carried out along \( a'' \) and \( c'' \), respectively. Conversely, as temperature is increased to \( k_B T \sim J \) this qualitative difference is wiped out. Now, for both directions a common paramagnetic magnetization response is observed.

Furthermore, for fields directed along \( a'' \) the singular dependence of the magnetization at \( T = 0 \) and \( H = H_{\text{sat}} \) on temperature and field becomes visible in the figure. However, already for values \( k_B T/J = 0.04 \) the zero temperature singular field dependence has disappeared in the experiments. In contrast, for a field directed along \( c'' \) such a behavior is not observed, reflecting that because of the locally induced staggered field being a relevant factor for this field direction the singularity is absent even at zero temperature.

For fields along \( c'' \) the influence of the inhomogeneity parameter \( k \) becomes evident. In Fig. 1(c) we compare the low temperature magnetization of Cu Pyrimidine \( (k = 0.11 \ [14]) \) and Cu Benzoate \( (k = 0.043 \ [15]) \). With the larger inhomogeneity parameter, for Cu Pyrimidine we observe a more pronounced non-linearity of \( M \) at low fields, while saturation is delayed at high fields. For further analysis, we have to take into account that the measured magnetization represents a superposition of longitudinal and transverse magnetization components, with the physically measured magnetization \( M_{\text{phys}} \) containing a staggered component \( M_{\text{staggered}} \) and a uniform one \( M_{\text{uniform}} \):

\[
M_{\text{phys}} = M_{\text{uniform}} + kM_{\text{staggered}}.
\] (4)

Figure 2. (a) The reduced staggered magnetization \( M_{\text{staggered}}/M_{\text{sat}} \) from exact diagonalization \( \text{(ED)} \) as function of reduced magnetic field \( \mu_0 H/\mu_0 H_{\text{sat}} \) for different values of the inhomogeneity parameter \( k \) at \( k_B T/J = 0 \). (b) \( M_{\text{staggered}}/M_{\text{sat}} \) from transfer matrix renormalization group analysis \( \text{(TMRG)} \) as function of \( \mu_0 H/\mu_0 H_{\text{sat}} \) for different values of \( k \) and \( k_B T/J \). For comparison, we include the ED calculations for \( k = 0.11 \); for details see text.
Correspondingly, in Fig. 2 we plot the calculated reduced staggered magnetization $M_{\text{staggered}}/M_{\text{sat}}$ as function of reduced magnetic field $\mu_0 H/\mu_0 H_{\text{sat}}$ for different values of the inhomogeneity parameter $k$. Calculations via exact diagonalization have been carried out for zero temperature and finite spin chains of $N = 20$ sites ($k = 0.08, 0.11$) and $16$ sites ($k = 0.16, 0.28$), respectively. The TMRG calculations have been performed at finite temperatures $k_B T/J = 0.09$ ($k = 0.043$) and $k_B T/J = 0.04$ ($k = 0.11$), respectively (for details see Refs. [14,15]).

The figure illustrates the sensitivity of the magnetization to staggering effects induced by small perturbations to the common Heisenberg Hamiltonian via DM interaction and $g$ tensor anisotropy. From the figure it can bee seen that a local staggered field $h_s$ only of the order of $10\%$ of the uniform applied field induces a staggered magnetization of the order of up to $70\%$ of the saturation magnetization. Moreover, from the figure the range of applicability of the exact diagonalization calculations can be identified. For the present calculations with up to $N = 20$ sites finite-size effects become relevant below $\mu_0 H/\mu_0 H_{\text{sat}} \sim 0.2$. In this field range, structure appears in the field dependence of the staggered magnetization. In contrast, no structure is observed for the corresponding TMRG calculation, which is free of finite-size effects by construction. Conversely, at higher fields finite-size effects can be ignored for the ED calculations, which aside from temperature effects yield a staggered magnetization in full agreement with the results from TMRG.

In conclusion, we present a combined experimental and theoretical study of the magnetization of the staggered $S = 1/2$ antiferromagnetic Heisenberg chain. We discuss the temperature and field dependence for the directions of purely uniform and staggered behavior, this in context of the quantum criticality of the uniform $S = 1/2$ antiferromagnetic Heisenberg chain. From the data we derive the field dependence of the staggered magnetization $M_{\text{staggered}}$ and discuss the dependence of $M_{\text{staggered}}$ on the inhomogeneity parameter $k$.

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