Magnetization Process of the S=1/2 Distorted Diamond Spin Chain with the Dzyaloshinsky-Moriya interaction

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Abstract.
The distorted diamond chain has attracted a lot of interest, since the compound Cu3(CO3)2(OH)2, so-called azurite, was revealed to be a good candidate for this system. Although several theoretical studies were performed, the strong anisotropy observed in the magnetization measurement of azurite has not been explained very well. To solve the mechanism of this anisotropy, we introduce the Dzyaloshinsky-Moriya (DM) interaction suitable for the crystal symmetry of azurite. The numerical diagonalization study on this realistic model indicates that the anisotropy in the experimental magnetization curve would be reproduced with sufficiently large DM interaction. We also propose a mechanism of the field-induced incommensurate order, which was observed above the 1/3 magnetization plateau by the NMR measurement.

1. Introduction
The $S=1/2$ distorted diamond chain is one of interesting frustrated low-dimensional magnets. Due to large quantum fluctuation and frustration, various anomalous quantum phenomena have been predicted by several theoretical works [1, 2, 3], particularly in the external magnetic field. The compound Cu3(CO3)2(OH)2, so-called azurite, is one of good candidates to realize this system [4]. The recent magnetization measurement indicated a large magnetization plateau at 1/3 of the saturation magnetization, as predicted by the theoretical work based on the density matrix renormalization group approach [1]. The observed magnetization curve of azurite, however, exhibited a large anisotropy, namely a quite different shape between the two directions of external field: parallel and perpendicular to the $c$-axis of azurite. The width of the 1/3 plateau is about 90% larger in the latter case than the former. Gu and Su [2], using the finite-temperature density matrix renormalization group approach, tried to explain the anisotropy of the magnetization process. But they introduced a ferromagnetic exchange and Ising-like coupling anisotropy a priori, which are not expected from the crystal structure of azurite. Thus it would be important to explain the mechanism of the anisotropic magnetization process, based on some realistic interactions. In addition the recent NMR measurement on azurite indicated an
incommensurate order in higher magnetic field region than the 1/3 plateau \cite{5}. In this paper, we try to reproduce the anisotropic magnetization curve by introducing the DM interaction. The observed field-induced incommensurate order is also explained by the $\eta$ inversion mechanism proposed in our previous work \cite{6}.

2. Model
The Hamiltonian of the $S = 1/2$ distorted diamond chain model is written by

\[ \mathcal{H} = \mathcal{H}_0 + \mathcal{H}_Z \]

\[ \mathcal{H}_0 = J_1 \sum_{j=1}^L (S_{3j-1} \cdot S_{3j} + S_{3j} \cdot S_{3j+1}) + J_2 \sum_{j=1}^L S_{3j+1} \cdot S_{3j+2} \]

\[ + J_3 \sum_{j=1}^L (S_{3j-2} \cdot S_{3j} + S_{3j} \cdot S_{3j+2}) \]  

\[ \mathcal{H}_Z = -H \sum_{l=1}^{3L} S^a_l. \]  

All the coupling constants are positive (antiferromagnetic). $H$ is the magnetic field and $a$ is $x$ or $z$. Since the crystal symmetry of azurite allows the DM interaction at $J_1$ and $J_3$ bonds, we assume the DM interactions described by the following Hamiltonian,

\[ \mathcal{H}_{DM} = iD_1 \sum_{j=1}^L (S^+_{3j-1}S^-_{3j} - S^-_{3j-1}S^+_{3j} + S^+_{3j}S^-_{3j+1} - S^-_{3j}S^+_{3j+1}) \]

\[ + iD_3 \sum_{j=1}^L (S^+_{3j-2}S^-_{3j} - S^-_{3j-2}S^+_{3j} + S^+_{3j}S^-_{3j+2} - S^-_{3j}S^+_{3j+2}). \]  

We assume that $\vec{D}_1$ and $\vec{D}_3$ are in the same direction along the $c$-axis, and the ratios $D_1/J_1$ and $D_3/J_3$ are common as $D_1/J_1 = D_3/J_3 = \alpha$, for simplicity.

3. Magnetization curve
Fitting the theoretical magnetization curve calculated by the density matrix renormalization group method to the observed one (averaged between the two directions), a realistic parameter set was estimated as $J_2/J_1 = 1.25$ and $J_3/J_1 = 0.45$ in our previous work \cite{4}. In order to reproduce the anisotropy between the two direction, we calculated the magnetization curves of the present model including the DM interaction for $J_2/J_1 = 1.25$, $J_3/J_1 = 0.45$ and $\alpha = 0.2$, using the numerical exact diagonalization for $L = 8$ (24 spins). In Figs. 2 (a) and (b) the calculated magnetization curves for (a)$\vec{H} \parallel x$ ($\vec{H} \perp \vec{D}$) and (b)$\vec{H} \parallel z$ ($\vec{H} \parallel \vec{D}$) are shown together with the experimental curves of azurite for (a)$\vec{H} \parallel c$-axis and (b)$\vec{H} \perp c$-axis, respectively. Figs. 2 (a) and (b) suggest that the calculated magnetization curves well reproduce the observed
anisotropic behavior. Even with larger $D_1$ and $D_3$ than the present values, it was difficult to quantitatively reproduce the observed large difference of the 1/3 plateau width. The plateaux at zero magnetization and at two-thirds of the saturation magnetization are associated with the spontaneous breaking of the translational symmetry [1]. Therefore, they are realized in the very restricted regions of the phase diagrams on the plane of coupling constants, and thus they have very narrow widths and may be fragile against the DM and temperature effects.

4. Field-induced incommensurate order

Finally, we consider the field-induced incommensurate order observed above the 1/3 plateau of azurite. It was discovered in the case of $H \perp c$-axis, correspond to the configuration $H \parallel \vec{D}$ in the present work. Thus we assume the U(1) symmetry and the gapless phase above the 1/3 plateau can be described by the Tomonaga-Luttinger liquid characterized by the spin correlation functions,

$$\langle S^x_0 S^x_r \rangle \sim (-1)^r r^{-\eta^x}, \quad (6)$$

$$\langle S^z_0 S^z_r \rangle - m^2 \sim \cos(2k_F r)r^{-\eta_z}. \quad (7)$$

In usual antiferromagnets the former spin correlation is stronger than the latter and the interchain interaction would stabilize the canted Néel order. However, if the latter spin correlation is stronger ($\eta$ inversion [7, 6]), the incommensurate order along $H$ would be realized. Neglecting the DM interaction for simplicity, we estimated the critical exponents $\eta^x$ and $\eta^z$ using the finite-size scaling based on the conformal field theory and the numerical exact diagonalization. The calculated exponents at half of the saturation magnetization for $J_2/J_1 = 1.25$ are plotted versus $J_3/J_1$ in Fig. 3. The product $\eta^x \eta^z$ is also plotted as a dotted-dashed curve. It indicates that the universal relation $\eta^x \eta^z = 1$ of the Tomonaga-Luttinger liquid is well satisfied and supports a validity of the present analysis. As can be seen from Fig.3, the $\eta$ inversion occurs in the region $J_3/J_1 > 0.38$, in which the realistic parameter for azurite $J_3/J_1 = 0.45$ is located. It suggests that the field-induced incommensurate order occurs about 1/2 of the saturation magnetization, which would explain the observed incommensurate order above the 1/3 plateau. It also confirm that the parameter set $J_2/J_1 = 1.25$ and $J_3/J_1 = 0.45$ which was proposed in our previous work [4] are suitable for azurite.
Figure 3. \( \eta^x \) (solid curve) and \( \eta^z \) (dashed curve) for \( J_2/J_1 = 1.25 \) at half of the saturation magnetization. The product \( \eta^x \eta^z \) is also shown by a dot-dashed curve.

5. Summary
The distorted diamond chain with the DM interaction was investigated by the numerical exact diagonalization. The magnetization curves for \( J_2/J_1 = 1.25 \), \( J_3/J_1 = 0.45 \) and \( \alpha = 0.2 \) fairly well reproduced the difference between the cases of \( \vec{H} \parallel c \)-axis and \( \vec{H} \perp c \)-axis. In addition we successfully explained the mechanism of the field-induced incommensurate order, which had been observed above the 1/3 magnetization plateau by the NMR measurement.

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