Field-Induced Incommensurate Order and Possible Supersolid in the S=1/2 Frustrated Diamond Chain

T Sakai\textsuperscript{1}, K Okamoto\textsuperscript{2} and T Tonegawa\textsuperscript{3}

\textsuperscript{1}Japan Atomic Energy Agency (JAEA), SPring-8, Hyogo 679-5148, and Department of Material Science, University of Hyogo, Kamigori, Hyogo 678-1297, Japan
\textsuperscript{2}Department of Physics, Tokyo Institute of Technology, Tokyo 152-8551, Japan
\textsuperscript{3}Department of Mechanical Engineering, Fukui University of Technology, Fukui 910-8505, Japan

E-mail: sakai@spring8.or.jp

Abstract.

The magnetization process of the distorted diamond chain is theoretically investigated using the numerical exact diagonalization and the density matrix renormalization group calculation. It is found that for some specified parameters the ground-state critical behavior is dominated by the incommensurate spin correlation parallel to the external magnetic field, around the 2/3 magnetization plateau. In the presence of interchain interaction it should be an incommensurate long-range order which possibly leads to a supersolid if coexisting with the usual transverse antiferromagnetic long-range order.

The $S = 1/2$ distorted diamond chain is one of interesting frustrated systems. The previous study using the numerical exact diagonalization and the level spectroscopy indicated that this system would exhibit magnetization plateaux at 1/3 and 2/3 of the saturation magnetization with sufficiently strong frustration [1]. In fact the 1/3 magnetization plateau was observed in the real compound Cu$_3$(CO$_3$)$_2$(OH)$_2$ so-called azurite [2]. In this paper, we propose a field-induced incommensurate order around the 2/3 plateau. It can be realized when the incommensurate spin correlation parallel to the external magnetic field is larger than the perpendicular antiferromagnetic one, so called $\eta$ inversion indicated later, in the presence of interchain interaction. Such a possibility was also discussed for several quasi-1D frustrated systems [3, 4]. We present some phase diagrams including a magnetization plateau, because the $\eta$ inversion should be accompanied by the 2/3 plateau.

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

\begin{align}
\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} \\
&\quad + 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_l^z.
\end{align}
All the coupling constants are positive (antiferromagnetic). $H$ is the magnetic field.

Applying an external magnetic field to the present system(1), except for gapped regions, namely the 1/3 and 2/3 magnetization plateaux, the gapless Tomonaga-Luttinger liquid phase is realized [5, 6]. It is characterized by the power-law decay of the spin correlation functions $\langle S_0^x S_r^x \rangle \sim (-1)^r r^{-\eta^x}$ and $\langle S_0^z S_r^z \rangle - m^2 \sim \cos(2k_F r) r^{-\eta^z}$, where $k_F$ is the Fermi momentum of the Tomonaga-Luttinger liquid, which is related to the magnetization $m \equiv \sum_j S_j^z / L$ as $k_F = \pi m$. The first spin correlation function describes an ordinary antiferromagnetic correlation perpendicular to $H$ and it would be the long-range canté Néel order with appropriate interladder interaction. The second one corresponds to an incommensurate spin correlation parallel to $H$. In usual antiferromagnets, $\eta^x < \eta^z$ is satisfied and the transverse antiferromagnetic correlation is dominant, irrespective of $H$. As a result, the canted Néel order would be observed with interladder interaction. In the presence of some frustrated interaction, however, recent numerical studies revealed that some one-dimensional systems can exhibit $\eta^x > \eta^z$ in some intermediate $H$ close to the magnetization plateau where the translational symmetry is spontaneously broken [3, 7, 8]. Namely, the incommensurate spin correlation parallel to $H$ can be dominant. In this case, the incommensurate order can occur in the quasi-one-dimensional frustrated systems [3]. The realization of $\eta^x > \eta^z$ is called “$\eta$ inversion”. Since the universal relation $\eta^x \eta^z = 1$ should be held for the Tomonaga-Luttinger liquid, the boundary value of $H$ for the $\eta$ inversion should be the field for which $\eta^x = \eta^z = 1$. On the other hand, the previous study to indicate the 2/3 magnetization plateau in the present model (1) suggested [1] that the phase boundary of the plateau should be given by $\eta^x = \eta^z = 1$. It implies that the present system should exhibit the $\eta$ inversion at least in the parameter region where the 2/3 plateau appears. According to the level spectroscopy analysis [1], the 2/3 magnetization plateau was revealed to occur for $J_1 = 1.0$, $J_2 = 0.8$ and $0.168 < J_3 < 0.380$. Thus we fix the parameters as $J_1 = 1.0$, $J_2 = 0.8$ and vary $J_3$ from 0 to 0.5 in this paper.

The critical exponents $\eta^x$ and $\eta^z$ can be calculated numerically by the size-scaling forms $\Delta_1 \sim v_x \eta^x \frac{1}{L}$ and $\Delta_{2k_F} \sim v_x \eta^z \frac{1}{L}$, where $\Delta_1$ is the spin gap, $\Delta_{2k_F}$ the $2k_F$ excitation gap and $v_x$ is the sound velocity [6]. Neglecting the size correction, the calculated $\eta^x$ and $\eta^z$ for $L = 8$ are plotted versus the magnetization $m$ in Figures 2 (a) for $J_3 = 0.1$ and (b) for $J_3 = 0.2$, respectively. They show that for $J_3 = 0.2$ it holds $\eta^x > \eta^z$, namely $\eta$ inversion occurs around $m/m_s \sim 2/3$ ($m_s$ is the saturation magnetization), while it does not for $J_3 = 0.1$.

In order to present the phase diagram in $m$-$J_3$ plane, we estimate the critical value of $m$ satisfying $\eta^x(m) = 1$, because the finite-size correction is smaller for $\eta^x$ than $\eta^z$ [9]. The function $\eta^x(m)$ of finite systems is a discrete one calculated only for $m = n/L$, where $n$ is integer. Thus we perform a suitable interpolation to determine the point for $\eta^x(m) = 1$. The results of the phase diagram calculated for $L = 8$ is shown in Fig. 3(a) for $J_1 = 1.0$ and $J_2 = 0.8$. It is shown that the $\eta$ inversion region appears around $m \sim 2/3$ and the 2/3 plateau divides the region into two parts. The edges of the plateau line were determined by the previous level spectroscopy analysis [1].

The $H - J_3$ phase diagram would be much useful to consider some realistic situations observed in experiments. A similar interpolation technique for the external magnetic field $H$ based on the finite-cluster calculation for $L = 12$ yields the $H - J_3$ phase diagram in Fig. 3(b) for $J_1 = 1.0$.
and $J_2 = 0.8$. The 2/3 plateau is indicated by the region surrounded by dashed curves and the $\eta$ inversion appears inside of solid curves except for the plateau.

The magnetization curves for $L = 24$ calculated by the density matrix renormalization group (DMRG) method are shown in Figs. 4 (a) for $J_3 = 0.1$ and (b) for $J_3 = 0.2$, respectively. The level spectroscopy analysis [1] convincingly indicated that the 2/3 plateau exists in the thermodynamic limit for $J_3 = 0.2$, while not for $J_3 = 0.1$, although it is much smaller than the 1/3 one. Thus the $\eta$ inversion should also appear for $J_3 = 0.2$, as shown in Fig.3 (b). Figs.4(a) and (b) suggest that it is difficult to identify the $\eta$ inversion in the shape of the magnetization curve, because it is not a phase transition, but a crossover in 1D. If interchain interactions exist, some anomalous behaviors would appear as a phase transition.

In summary, the $S = 1/2$ distorted diamond chain in magnetic field is investigated by the numerical exact diagonalization and DMRG calculations. The present study indicates that, instead of usual transverse antiferromagnetic spin correlation, the incommensurate one parallel to $H$ can be dominant ($\eta$ inversion) around 2/3 of the saturation magnetization for some realistic parameters. Several phase diagrams including the plateau phase and the $\eta$ inversion regions

**Figure 2.** $\eta^x$ and $\eta^z$ for (a) $J_3 = 0.1$ and (b) $J_3 = 0.2$.

**Figure 3.** Phase diagrams in the (a) $m$-$J_3$ and (b) $H$-$J_3$ planes for $J_1 = 1.0$ and $J_2 = 0.8$. 
are presented. With appropriate interchain interaction, the incommensurate long-range order would appear in the \( \eta \) inversion region. A DMRG analysis with a mean field approximation for interchain interaction [3], suggested a first-order phase transition between the field-induced incommensurate and usual antiferromagnetic orders in the \( S = 1/2 \) frustrated bond-alternating chain. More detailed analyses, however, would possibly reveal the coexistence of the two orders at lower temperature phases. A field theoretical study [10] would indicate that these two ordered phases are results coming from the Bose-Einstein condensation of two different components, and the coexisting phase corresponds to the supersolid phase observed in \(^4\)He [11]. A realistic parameter set for the azurite estimated from the magnetization curve [2] is not in the range of the \( \eta \) inversion, but it is still controversial [12]. Actually an incommensurate spin correlation was observed at an intermediate field region between the 1/3 and 2/3 plateaux in the recent NMR measurement on the azurite [13]. Further detailed measurements around the 2/3 plateau would possibly clarify the incommensurate phase which is predicted in this work.

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