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2006 J. Phys.: Condens. Matter 18 4819

(http://iopscience.iop.org/0953-8984/18/20/006)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **18** (2006) 4819–4826 doi:10.1088/0953-8984/18/20/006

Possibility of field-induced incommensurate order in a quasi-one-dimensional frustrated spin system

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Received 20 January 2006
Published 2 May 2006
Online at stacks.iop.org/JPhysCM/18/4819

Abstract
We study an incommensurate long-range order induced by an external magnetic field in a quasi-one-dimensional bond-alternating spin system, F_5PNN, focusing on the role of the frustrating interaction which can be enhanced by a high-pressure effect. On the basis of the density matrix renormalization group analysis of a microscopic model for F_5PNN, we present several H–T phase diagrams for typical parameters of the frustrating next-nearest-neighbour coupling and the interchain interaction, and then discuss how the field-induced incommensurate order develops by the frustration effect in such phase diagrams. A magnetization plateau at half the saturation moment is also mentioned.

1. Introduction

Quasi-one-dimensional quantum spin systems exhibit several interesting field-induced phenomena. In a gapped spin chain an external magnetic field exhibits a quantum phase transition to the gapless Tomonaga–Luttinger liquid phase at some critical field H_{c1} [1, 2]. The Tomonaga–Luttinger liquid phase basically continues up to the saturation field H_{c2} and is characterized by the power-law decay of the spin correlation function:

$$\langle S_0^x S_r^x \rangle \sim (-1)^r r^{-\eta_x}, \tag{1}$$

$$\langle S_0^z S_r^z \rangle - m^2 \sim \cos(2k_F r) r^{-\eta_z}, \tag{2}$$

where m is the magnetization along the field $H \parallel z$, and the Fermi wavenumber k_F depends on the magnetization. Since these exponents satisfy $\eta_x < \eta_z$ in usual antiferromagnets, the commensurate antiferromagnetic long-range order perpendicular to H should appear in the presence of interchain interactions. In fact, such a field-induced antiferromagnetic order has been theoretically proposed [3] and experimentally observed in some quasi-one-dimensional gapped systems [4, 5]. Here it should be noted that the ordered phase can also be interpreted as a result of the magnon Bose–Einstein condensation [6].

In some one-dimensional (1D) frustrated systems, however, the η-inversion, namely $\eta_x > \eta_z$, has been predicted for some intermediate strength of the magnetic field between H_{c1} and H_{c2} [7–11]. Indeed, such η-inversion has been suggested experimentally by the NMR relaxation rate $1/T_1$ of an organic $S = 1/2$ bond-alternating chain, pentafluorophenyl nitronyl nitroxide (F_5PNN), in a certain temperature region where a power-law behaviour of $1/T_1$ can be observed [12, 13]. A more interesting point associated with the η-inversion is that the field-induced long-range *incommensurate order* can appear, reflecting the enhancement of the incommensurate correlation [14]. Although the recent specific heat measurement [15, 16] indicates that the field-induced commensurate antiferromagnetic order may be realized for F_5PNN, the NMR experiments are still suggesting an enhancement of the incommensurate correlation in F_5PNN. Thus, it seems that F_5PNN is located in the vicinity of the boundary between the commensurate order phase and the incommensurate order phase. In this paper, we discuss the possibility of realizing the field-induced incommensurate order in F_5PNN, putting a special emphasis on a high-pressure effect which can enhance the frustrating interaction. We examine how the incommensurate order appears in the phase diagram on the basis of density matrix renormalization group calculations of a microscopic model for F_5PNN. We also mention the possibility of a magnetization plateau and coexistence of the commensurate and incommensurate orders.

2. Model

The experimentally observed η-inversion and the field-dependent crossover of the bond-alternating ratio [17] have indicated the presence of frustration in F_5PNN. Thus, we consider the bond-alternating spin chain with the next-nearest-neighbour interaction in a magnetic field

$$\hat{H} = J_1 \sum_j \vec{S}_{2j} \cdot \vec{S}_{2j+1} + J_2 \sum_j \vec{S}_{2j+1} \cdot \vec{S}_{2j+2} \tag{3}$$

$$+ J' \sum_j \vec{S}_{2j} \cdot \vec{S}_{2j+2} - H \sum_j S^z_j, \tag{4}$$

as a realistic model for F_5PNN, where J_1 and J_2 ($\neq J_1$) denote the alternating nearest-neighbour interactions, and J' means the next-nearest-neighbour coupling. In what follows, J_1 is set to unity for simplicity. For $H_{c1} < H < H_{c2}$, the model is basically in the Tomonaga–Luttinger liquid phase with the Fermi wavenumber $k_F = (1/2 - m)\pi$.

3. Mechanism of the field-induced incommensurate order

Let us briefly explain the mechanism of the field-induced incommensurate order. If J' is sufficiently large, the η-inversion occurs around $m \sim m_s/2$, where m_s is the saturated magnetization [8, 9, 14]. At the magnetic field in the η-inversion region, the dominant spin correlation should not be the usual commensurate antiferromagnetic one perpendicular to H, but the incommensurate one along the H-direction. In the presence of interchain interactions, the long-range order corresponding to the dominant spin correlation is expected, implying that a long-range incommensurate order in the H direction can be realized for the region $\eta_x > \eta_z$.

In the present study, the dominant spin correlation is determined by two staggered susceptibilities $\chi_\perp$ and $\chi_\parallel$ corresponding to the spin correlations (1) and (2), respectively.

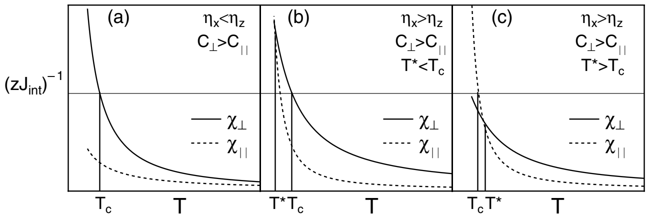

Figure 1. Schematic temperature dependence of $\chi_\perp$ and $\chi_\parallel$ in three cases. At $T = T_c$, the larger susceptibility satisfies the relation (6). At $T = T^*$ in (b) and (c), $\chi_\perp$ is equal to $\chi_\parallel$.

Low temperature behaviours of the susceptibilities are characterized by the exponents η_x and η_z [10]:

$$\chi_\perp(T) \sim C_\perp T^{-(2-\eta_x)} \qquad \text{and} \qquad \chi_\parallel(T) \sim C_\parallel T^{-(2-\eta_z)}, \tag{5}$$

where $C_\perp$ and $C_\parallel$ are nonuniversal coefficients. Equation (5) shows that the more dominant spin correlation corresponds to the more strongly divergent susceptibility. When the η-inversion occurs, for example, $\chi_\parallel$ shows stronger divergence than $\chi_\perp$.

For quantitative treatment of the field-induced order and of the accompanying phase transition, we use the above susceptibilities. Based on the mean-field approximation for the interchain interaction J_{int} [18], the critical temperature T_c is given by

$$\chi_\gamma(T_c) = (zJ_{\text{int}})^{-1}, \tag{6}$$

where γ is $\perp$ or $\parallel$, and z the number of adjacent chains.

On the basis of equations (5) and (6), we can classify possible types of field-induced orders; we consider the following cases, which are depicted in figure 1. The first one is that $\eta_x < \eta_z$ (figure 1(a))[7]. In this case, since $\chi_\perp$ strongly diverges, $\chi_\perp > \chi_\parallel$ is always realized. As T decreases, $\chi_\perp$ first satisfies the relation (6) and the commensurate order appears below T_c. By contrast, if the η-inversion occurs (i.e. $\eta_x > \eta_z$), $\chi_\perp < \chi_\parallel$ is naively expected. However, we should pay attention to the magnitude of the nonuniversal coefficients $C_\perp$ and $C_\parallel$. We have found that, even for $\eta_x > \eta_z$, $C_\perp$ is larger than $C_\parallel$, and thus $\chi_\perp$ can be larger than $\chi_\parallel$ at high temperatures, although $\chi_\parallel$ finally exceeds $\chi_\perp$ at $T = T^*$. If $T_c > T^*$, therefore, $\chi_\perp$ satisfies equation (6) at a higher temperature and the commensurate order appears (see figure 1(b)). In such a case, another phase transition is expected to occur from the commensurate phase to the incommensurate phase at $T = T^*$. The previous DMRG analysis of the free energy has illustrated that the transition actually occurs when zJ_{int} is not so large [19]. For $T_c < T^*$ (figure 1(c)), $\chi_\parallel$ satisfies equation (6) first and only the incommensurate order appears.

4. Phase diagrams

By a numerical calculation of $\chi_\perp$ and $\chi_\parallel$ for a single chain and the mean-field treatment described above, we have obtained H–T phase diagrams. For the calculation at finite temperature in the thermodynamic limit, we use the finite temperature DMRG [20, 21]. The DMRG calculation has been carried out with the maximum number of retained bases $m = 64$ and the truncation error is at most of the order of 10^{-6}. Technical details to obtain $\chi_\perp$ and $\chi_\parallel$ are described in [14].

In figure 2 we show the phase diagrams for three typical parameters of the frustrating coupling: (a) $J' = 0.05$ (small J' case), (b) $J' = 0.1$ (intermediate J' case), and (c) $J' = 0.15$

[7] For $\eta_x < \eta_z$, $C_\perp > C_\parallel$ is usually satisfied, where the frustration effect is not significant.

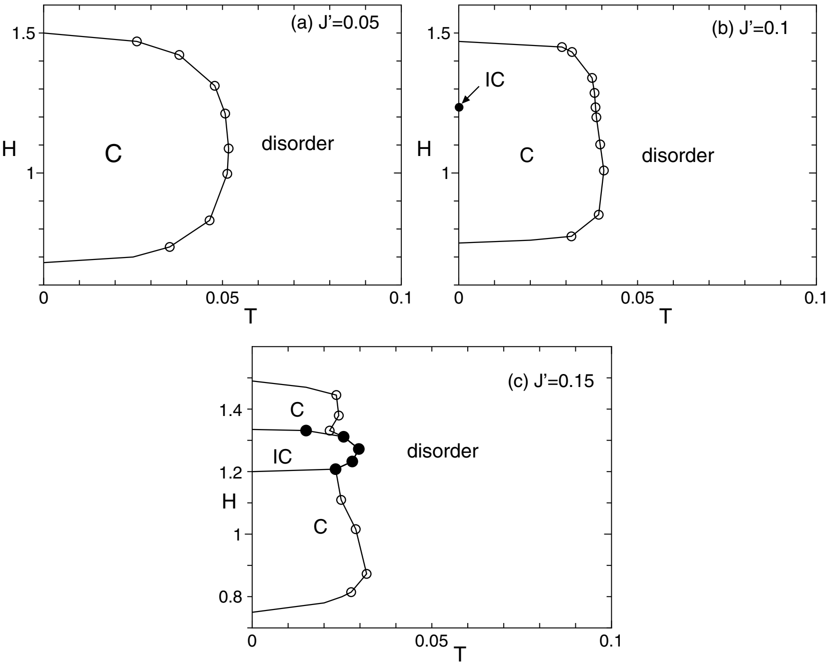

Figure 2. H–T phase diagram for various J' ($J' = 0.05$, 0.1 and 0.15). C and IC denote the commensurate antiferromagnetic and incommensurate order phases, respectively. Filled circles denote the transition temperature between the IC phase and other phases, and open circles indicate that between the C phase and the disordered phase. The interchain interaction is fixed to $zJ_{\mathrm{int}} = 1/12$.

(large J' case). The interchain interaction is fixed to $zJ_{\mathrm{int}} = 1/12$, and J_2 is fixed to a realistic value 0.45 for F_5PNN [14]. In the figure, C and IC denote the commensurate antiferromagnetic-order phase and the incommensurate one, respectively.

4.1. Case (a)

The phase diagram in case (a) is typical for spin-gapped quasi-1D spin systems, where it should be remarked that the shape of the phase boundary between the C phase and the disordered phase looks like a semicircle. Since $\chi_\perp$ is always larger than $\chi_\parallel$ as shown in figure 1(a), the IC phase never appears.

4.2. Case (b)

In case (b), the phase diagram is clearly different from case (a) in the following two points. The first one is the appearance of the IC phase in a very low temperature region. At $H = 1.23$, where the magnetization m at $T = 0$ is equal to $m_s/2$, the η-inversion occurs and thus $\chi_\parallel$ shows slightly stronger divergence than that of $\chi_\perp$. Thus the IC phase can appear around $H = 1.23$ in the quite low temperature region ($T < T^*$). However, in the high temperature region ($T > T^*$), $\chi_\perp$ is larger than $\chi_\parallel$, resulting in the transition between the commensurate order phase and the disordered phase at $T = T_c$. These behaviours of the staggered susceptibilities correspond to figure 1(b). Another important point is that the phase boundary between the C phase and the disordered phase is deformed from the semicircle-shape, particularly around $H = 1.23$. This

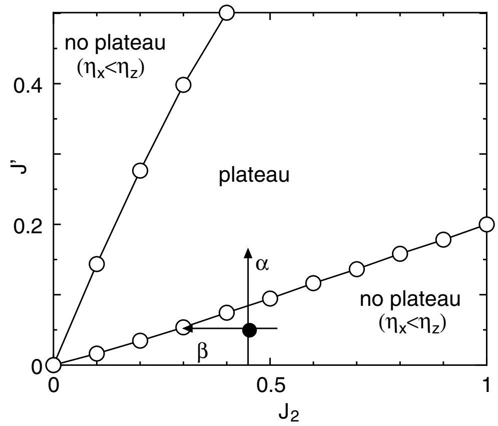

Figure 3. J_2–J' phase diagram of the dominant correlations at $m = m_s/2$ at zero temperature, which is equivalent to the 1/2 plateau phase diagram in [25]. The filled circle corresponds to the parameters of F_5PNN.

is because the transverse fluctuation is suppressed by the frustration effect even in $T > T^*$, resulting in the lowering of T_c.

4.3. Case (c)

In case (c), the large J' enhances the incommensurate correlation and extends the region of the IC phase. Around $H = 1.27$, $\chi_\perp$ behaves as in figure 1(c) and thus we can observe the direct transition from the disordered phase to the IC phase, which divides the C phase into the low-H part and the high-H part.

5. Discussion

In the previous work [14], the authors estimated J' of F_5PNN as ∼0.05 (=0.3 K), based on a qualitative analysis of the magnetization curve. This value is slightly smaller than the critical value (∼0.08) for the η-inversion or the emergence of the incommensurate order (see figure 3). We think however that high-pressure experiments can realize the incommensurate phase within realistic experimental situations. For example, a hydrostatic pressure experiment reveals a drastic change of the bond-alternation ratio in powder samples of F_5PNN [22]. In addition, a recent experiment clarifies that specific heat of the power samples has slightly different temperature dependence from that of a single crystal of F_5PNN, which can be attributed to an effective pressure caused by an experimentally technical reason [23]. These experimental results strongly indicate that the properties of F_5PNN are quite sensitive to the pressure. Thus it is worthwhile to discuss precisely how the incommensurate order phase develops in the H–T phase diagram near the phase boundary of the η-inversion.

As in figure 3, we can readily consider two possible paths such that the couplings of F_5PNN exceed the critical value for the incommensurate order: increasing the next-nearest neighbour interaction J' (arrow α) and/or decreasing the alternating coupling J_2 (arrow β). Here, we chiefly discuss the J'-dependence of the H–T phase diagrams along the vertical arrow direction in figure 3, assuming that $J_{\rm int}$ is independent of the pressure. We can then expect that the features of H–T phase diagrams should change as figures 2(a)–(c). If J' is sufficiently enhanced by the pressure, the direct phase transition from the disordered phase to

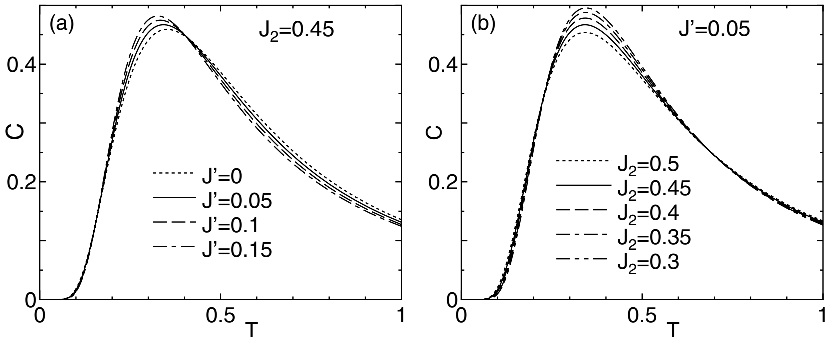

Figure 4. Temperature dependence of the specific heat for various values of exchange parameters. Each figure corresponds to the case where the exchange interactions are varied along the arrows α and β in figure 3.

the IC phase would be observed in a middle range of the magnetic field as in case (c). As mentioned in the previous section, however, a notable point is that, even in case (b), we may find the deformation of the phase boundary between the disordered phase and the C phase. From the experimental point of view, such a deformation of the phase boundary can be an important signal of a development of the IC order induced by the frustration, though it may be difficult to capture the IC phase at a very low temperature.

Next, we discuss how the changes of the exchange interactions can reflect on a measurement of the specific heat at *the zero magnetic field*, which is quite helpful in analysis of the experimental results, because it is not an easy work to obtain a complete H–T phase diagram in actual experiments [15, 16]. Figure 4 shows the temperature dependence of the specific heat for various values of J' and J_2 along the arrows in figure 3. When the frustrating interaction J' increases along the α direction, it is found that the pronounced peak around $T = 0.3$ sharpens. We note that this can be attributed to the increase of the density of states caused by the narrowing of the $S = 1$ magnon band width [24]. On the other hand, such a band narrowing also occurs as J_2 is reduced along the β direction. We can then see that the peak sharpens as well. We therefore think that a careful analysis of the shape of the specific heat peak at $H = 0$ provides important information for the enhancement of the IC correlation in the intermediate magnetic field region, although the pressure effect subtly influences both J' and J_2 in the actual situation.

Here, we should make a comment on the pressure effect for the *interchain* interaction. If $J_{\rm int}$ is also enhanced by the pressure, the direct phase transition from the disordered phase to the IC phase may be suppressed within the interchain mean-field analysis as shown in figure 5, where the interchain interaction is assumed to be unfrustrating. Nevertheless, the precise analysis of the crystal structure [17] of F_5PNN has indicated that the interchain couplings are also frustrating, which would effectively decrease value of $J_{\rm int}$ in the mean-field approximation. In order to reveal the role of the frustrating interchain interaction, of course, a precise analysis beyond the mean-field level is clearly required.

Finally, we want to discuss a magnetization plateau at half of the full moment. As in figure 3, the 1/2 plateau is closely related to the mechanism of the η-inversion; the criterion for η-inversion is equivalent to the one for plateau formation [14]. The level spectroscopy analysis [25] on model (4) has indicated the 1/2 plateau should appear at half the saturated magnetization for $J_2 = 0.45$ and $J' = 0.15$. However, we think that it may be difficult to observe clear evidence of the 1/2 plateau of F_5PNN even with a high-pressure experiment.

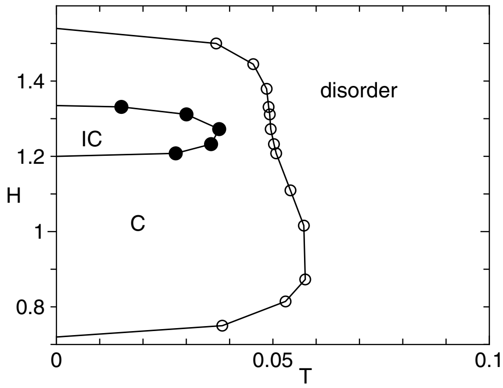

Figure 5. H–T phase diagram in case of large J' and large J_{int} ($J' = 0.15$ and $zJ_{\mathrm{int}} = 1/6$).

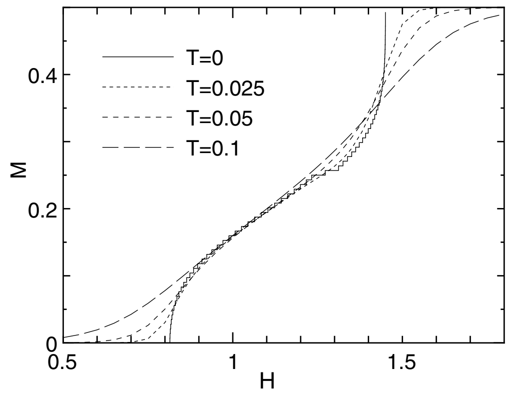

Figure 6. Magnetization curves for the realistic parameters $J_2 = 0.45$ and $J' = 0.15$ at several temperatures.

This is because the width of the plateau is still very narrow for these couplings, and thus the thermal fluctuation smears out the plateau even at temperatures where we can observe the magnetization. In figure 6, we show the magnetization curves for $J_2 = 0.45$ and $J' = 0.15$ obtained by the finite temperature DMRG, illustrating that the plateau is easily smeared out by the thermal effect. We think that, as for F_5PNN, it is more hopeful to verify the presence of the IC order than to make a direct observation of the $1/2$ plateau.

To summarize, we have discussed field-induced IC order for the bond-alternating spin chain with the next-nearest interaction, which is relevant to the high-pressure experiments of F_5PNN. In particular, we have analysed how the various physical quantities behave near the phase boundary, and then concluded that the precise analysis for the phase boundary and the zero-field specific heat can detect a signal of the field-induced IC order. Further experimental studies on F_5PNN are quite interesting. In addition, it is also a challenging issue to search for other spin chains exhibiting field-induced incommensurate order. A recently observed new phase of NDMAP in magnetic field might be a possible candidate [26].

The field-induced commensurate order in a quantum spin system is often described as the Bose–Einstein condensation (BEC) of a magnon gas. The present IC order may be viewed as a result of a 'charge order' of the magnon. As mentioned in [19], the transition between the field-induced incommensurate order and the usual commensurate antiferromagnetic one is of the first order. Then we can expect the coexistence of the two orders at the phase boundary. Recently, such a coexistence of the two different orders has actually been observed in solid ^{4}He, which is called a 'super-solid' state [27]. A connection to such an exotic state is also an interesting problem both from experimental and theoretical viewpoints.

Acknowledgments

We wish to thank Professors I Affleck, N Prokof'ev, S Suga, T Goto, Drs K Izumi, T Suzuki and Y Yoshida for fruitful discussions. This work is partly supported by Grants-in-Aid for Scientific Research (B) (No 17340100, No 14340099), (C) (No 14540329, No 16540332, No 17540317), and a Grant-in-Aid for Scientific Research on Priority Areas 'High field spin science in 100T' (No 451) from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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