Field-induced quantum ordered phases in an anisotropic Haldane system

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Abstract. Field-induced quantum ordered phases in the anisotropic $S = 1$ Haldane system are studied, using an effective Lagrangian formalism. We predict that, when the applied field is inclined from the principal axes of the spin anisotropy, successive phase transitions occur from the Haldane phase to a field-induced ordered phase followed by another spin-reorientation transition from the one to other field-induced ordered phase at high fields. Depending on the anisotropy constants, the spin-reorientation transition is of the first order or of the second order via another type of the ordered phase. Relevant implications of these transitions are further discussed in connection with the novel ordered phase recently observed in the Haldane system, NDMAP, at high fields.

1. Introduction
Field-induced quantum ordered phases in various low-dimensional magnetic chains have attracted much interest recently. These are the consequence of the applied magnetic field controlling quantum fluctuations, which play a particularly important role in one-dimensional antiferromagnetic spin systems and sometimes destroy the antiferromagnetic ordering. The resultant ground state is singlet, such as the Haldane state in an $S = 1$ spin chain with nearest neighbor exchange interactions.

In order to understand such a ground state, many researchers are interested in the recovery of magnetism and the corresponding magnetic ordering, destroying the gap by a strong external magnetic field. In fact, such a phase transition has been first realized experimentally in the Haldane system, $\text{Ni(C}_5\text{H}_{14}\text{N}_2)\text{_2N}_3\text{(PF}_6)$, abbreviated hereafter as NDMAP[1, 2], and provided us various interesting ingredients.

It is, however, noted that little attention has been focused on the quantum fluctuation in the quantum ordered phases. Although the quantum fluctuation is reduced by the order, it may still play a vital role and in some cases may lead to a phase transition between the ordered phases. To discuss these, it is crucial to consider the direction of the magnetic field relative to the principal axes of the spin anisotropy.

In this paper, we investigate the role of the quantum fluctuation in the field-induced quantum ordered phases and discuss the phase transition between the quantum ordered phases, motivated by the recent observation of the successive phase transitions in NDMAP[3]. This is a consequence of the competition among the exchange energy, the Zeeman energy and the anisotropy energy. Then, the direction of the field with respect to the principal axes of the anisotropy is one of the key ingredients[4].
In order to obtain the unified understanding of the phenomena, we use in this paper the phenomenological field theory (PFT) consisting of the effective Lagrangian, which based on the following Hamiltonian describing the anisotropic Haldane system under the external magnetic field\[5, 6, 7\],

$$
\mathcal{H}/J = \sum_i \left[ S_i \cdot S_{i+1} + d(S_i^z)^2 - e\{(S_i^x)^2 - (S_i^y)^2\} - h \cdot S_i \right],
$$

(1)

where \( S \) is the \( S = 1 \) spin operator and the Cartesian coordinates \((x, y, z)\) are referred to the spin anisotropy axes. The parameters, \( J, d, e \) and \( h \) denote, respectively, the exchange constant, the easy-plane anisotropy constant \( d \), the in-plane anisotropy constant \( e \) and the external magnetic field \( h = h(\sin \theta, 0, \cos \theta) \).

The present paper is organized as follows. In the next section, we describe the model Lagrangian. In \( \S \)3, we present theoretical results and discuss possible implications of the present results in connection with the experimental results in NDMAP. The last section is devoted to our concluding remarks.

2. Model Lagrangian

Using a Landau-Ginsburg type field theory, we follow the Lagrangian developed by Zheludev et al.\[8\], extending so that it can describe the anisotropy terms in the higher order:

$$
\mathcal{L} = \sum_{i=x,y,z} \left\{ \frac{1}{m_i} \left[ (\partial_t \phi_i)^2 - v_i^2 (\partial_x \phi_i)^2 \right] - 2 \frac{1}{m_i} (h \times \phi)_i \partial_t \phi_i \right\} - U_2 (\phi) - U_4 (\phi, \partial_t \phi),
$$

(2)

where \( \phi \) and \( v \) denote, respectively, the staggered order parameter, which is related to the sublattice magnetization, and the characteristic velocity. The quadratic and quartic parts of the potential are given by

$$
U_2 (\phi) = \sum_i \left[ m_i \phi_i^2 - \frac{1}{m_i} (h \times \phi)_i^2 \right],
$$

(3)

$$
U_4 (\phi, \partial_t \phi) = \sum_i \lambda_i \phi_i^2 \phi_i^2 + \sum_{ij} \lambda_{1,ij} \phi_i \phi_j \frac{1}{m_i m_j} F_i^2 + \sum_{ij} \lambda_{2,ij} \frac{\phi_i \phi_j}{m_i m_j} F_i F_j,
$$

(4)

$$
F = -\partial_t \phi + h \times \phi.
$$

(5)

Note that \( \lambda_{2,ij} \) is a symmetric tensor, \( \lambda_{2,ij} = \lambda_{2,ji} \), since \( \partial_{\phi_i} (\partial_{\phi_j} U_4) = \partial_{\phi_j} (\partial_{\phi_i} U_4) \).

In order to study the ground state of the system, we look for a minimum of the potential in the three dimensional \( \phi \) space: there are possibilities to have solutions (i) \( \phi = 0 \) or (ii) \( \phi = \phi^{(0)} \), where \( \phi^{(0)} \) has one component, two components or three components. The latter three states are the ordered states.

Once the ground state is determined, we can calculate excitation spectra using the Lagrange equation of motion, where the small fluctuations around the static solution are treated up to quadratic order. Thus, three excitation modes are obtained as functions of the field.

3. Results and discussions

We present results for the two sets of parameters listed in Table 1: The case (i) yields the first order phase transition while the case (ii) yields the second order phase transitions.

3.1. First order phase transition between two kinds of ordered phases

The phenomenological parameters, \( m, m_i, \lambda, \lambda_1 \), and \( \lambda_2 \), are determined so that PFT reproduces the excitation spectrum as a function of the field along the \( a \)-axis and the \( c \)-axis, observed in NDMAP by the neutron inelastic scattering (NIS)[10] and by the electron spin resonance
Table 1. Two sets of the phenomenological parameters used in the calculations.

| Case (i) | Case (ii) |
|----------|-----------|
| $m_x = 0.19$, $m_y = 0.29$, $m_z = 1.7$ | $\tilde{m}_x = 0.15$, $\tilde{m}_y = 0.15$, $\tilde{m}_z = 0.32$ |
| $\lambda = (1.4 \ 1.0 \ 1.0)$ | $\lambda = (1.2 \ 1.0 \ 1.0)$ |
| $\lambda_1 = (0.37 \ 0.37 \ 0.37)$ | $\lambda_1 = (0.37 \ 0.37 \ 0.37)$ |
| $\lambda_2 = (0.30 \ 0.30 \ 0.49)$ | $\lambda_2 = (0.30 \ 0.30 \ 0.62)$ |
| $\lambda_2 = (0.49 \ 0.44 \ 0.30)$ | $\lambda_2 = (0.62 \ 0.44 \ 0.30)$ |

(ESR)[9]: see the case (i) in Table 1. Note that these parameters are not unique. In Fig. 1(a), we show the excitation energies calculated as a function of the field along the $c$-axis.

In Fig. 1(c), the phase diagram is shown. Let us consider the case where $\theta = 0.1\pi$, which corresponds to the case where the magnetic field is applied along the $c$-axis of NDMAP. With increasing the field, the Haldane phase becomes unstable against the $xz$-ordered phase and eventually changes to it by the second order transition. Increasing further the field, there occurs a first order phase transition from the $xz$-ordered phase to the $y$-ordered phase. In between, both phases coexist. These facts are seen in Fig. 1(b), where the order parameters are presented as a function of the field. The latter phase transition between two kinds of the ordered phases can be said to be “spin-reorientation transition”. These results are qualitatively consistent with the results of our ab initio simulation[11] in which the density matrix renormalization group method is used.

3.2. Second order phase transition through another ordered phase

When we choose another set of parameters, the case (ii) in Table 1, we have a different situation in the phase diagram shown in Fig. 2. Although the Haldane phase, the $xz$-ordered phase and

Figure 1. The excitation energies (a) and the order parameter (b) are plotted as a function of the magnetic field along the $c$-axis. In (c) the phase diagram is plotted in the field $h$ versus the angle of the field $\theta/\pi$. These are the results calculated using the parameter set (i) in Table 1.
Figure 2. The excitation energies (a) and the order parameter (b) are plotted as a function of the magnetic field for $\theta/\pi = 0.1$. In (c) the phase diagram is plotted in the field $h$ versus the angle of the field $\theta/\pi$. These are the results calculated using the parameter set (ii) in Table 1.

the y-ordered phase share the $h$ vs $\theta/\pi$ plane as in the case (i), the spin reorientation transitions are of the second order in this case. Between the xz-ordered phase and the y-ordered phase, there appears another phase called the xyz-ordered phase, which has three components and hence this phase connects smoothly from the xz-ordered phase to the y-ordered phase. All spin-reorientation transitions are of the second order.

4. Concluding remarks
We have investigated the phase diagram of the anisotropic $S = 1$ Haldane system, using an effective Lagrangian formalism. It is interesting to mention that, when the applied field is inclined from the principal axes of the spin anisotropy, the successive phase transitions occur from the Haldane phase to a field-induced ordered phase followed by another spin-reorientation transition from the one to other field-induced ordered phase at high fields. This finding is favorably compared with the phase transition observed in NDMAP, although more sophisticated theories as well as experiments are still needed for quantitative discussions.

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