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Long-range order in the ground state of the $S = 1$ isotropic bilinear–biquadratic exchange Hamiltonian

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Abstract
We apply the method of infrared bounds with the technique of Kennedy, Lieb and Shastry to the isotropic spin-1 Hamiltonian with bilinear $(-J)$ and biquadratic $(-J')$ exchange interactions to examine the existence of antiferro-dipole long-range order (DLRO) and ferro-quadrupole long-range order (QLRO). We prove that DLRO exists at zero temperature in two and three dimensions for $0 \leqslant J' < -0.188J$ and $0 \leqslant J' < -1.954J$, respectively. In three dimensions we also prove the existence of QLRO in the ground state for $0 < 2J \leqslant J' < 2.664J$.

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1. Introduction

In various materials it has been recognized that quadrupole interactions induce quadrupole phase transitions; for example, $PrCu_2$ and CeAg are known to exhibit ferro-quadrupole (FQ) ordering (the cooperative Jahn–Teller phase transition) [1, 2] and CeB_6 and $PrPb_3$ antiferro-quadrupole ordering [3–5]. A variety of theoretical models have been proposed to study the nature of quadrupole systems [3, 4, 6–8]. One of the simplest models is described by the $S = 1$ Hamiltonian, which contains the isotropic biquadratic interaction in addition to the usual Heisenberg (bilinear) interaction:

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \boldsymbol{S}(i) \cdot \boldsymbol{S}(j) - J' \sum_{\langle i,j \rangle} (\boldsymbol{S}(i) \cdot \boldsymbol{S}(j))^2. \tag{1}$$

In this paper we are concerned with Hamiltonian (1) on the hypercubic lattice, and in particular we investigate the existence of long-range order (LRO) in the ground state of this Hamiltonian.

In the case of one dimension, Hamiltonian (1) has been studied extensively following the Haldane conjecture. There are a number of theoretical, numerical and experimental works, and even a few rigorous results. The ground-state phase diagram expected to be valid in one dimension is summarized as follows. The region $-\pi/4 < \theta < \pi/4$ $(-J/\sqrt{J^2 + J'^2} = \cos\theta, -J'/\sqrt{J^2 + J'^2} = \sin\theta)$ is the Haldane phase, $\pi/4 < \theta < \pi/2$

the massless phase, $\pi/2 < \theta < 5\pi/4$ the ferromagnetic phase and $5\pi/4 < \theta < 7\pi/4$ the dimer phase. (See, e.g., [9–12] and references therein.)

On the other hand, in two or more dimensions, the phase diagram of the ground state of Hamiltonian (1) has not yet been understood sufficiently, except the region $\pi/2 < \theta < 5\pi/4$, in which the ground state is proved to be ferromagnetic [13]. The models have been studied mostly by using approximation methods such as a molecular field theory, a spin wave theory and a high-temperature series expansion [14–20]. According to these approximation theories, the region $-\pi/2 < \theta < 0$ is the antiferromagnetic (AF) phase, $-3\pi/4 < \theta < -\pi/2$ the ferro-quadrupole phase and $0 < \theta < \pi/2$ is expected to be the antiferro-quadrupole or the canted antiferromagnetic phase. Recently, Harada and Kawashima investigated the ground state of the two-dimensional system for $-\pi \leqslant \theta \leqslant 0$ by quantum Monte Carlo simulation [21]. Their results also suggested that the region $-\pi/2 < \theta \leqslant 0$ is the antiferromagnetic phase and that there is neither magnetization nor staggered magnetization for $-3\pi/4 < \theta < -\pi/2$. It is to be noted that there has been no rigorous result about the existence of the phase or LRO predicted by approximation and numerical methods.

In the present paper we focus our attention on the region $-3\pi/4 \leqslant \theta \leqslant 0(\theta \neq -\pi/2)$, which corresponds to $J' \geqslant J, J' \geqslant 0(J \neq 0)$. In this region, as mentioned above, it is expected that antiferro-dipole long-range order (DLRO) or ferro-quadrupole long-range order (QLRO) exists in two or more dimensions.

Our purpose is to establish the existence of LRO in the infinite system; but here it is worth noting the rigorous results in the finite systems. When the system size is finite and the number of sites is even, by using the Marshall–Lieb–Mattis argument, it is proved that the ground state of Hamiltonian (1) in the region $J' > J, J' \geqslant 0(J \neq 0)$ is unique [22–24]. For $J < 0, J' \geqslant 0$ the unique ground state satisfies the so-called Marshall sign rule. It is also known that the antiferro-dipole correlation function for this ground state is bounded away from zero. (It is noted that this fact does not mean the existence of DLRO in the infinite systems.) On the other hand, for $J' > J > 0$ the unique ground state satisfies the 'Parkinson sign rule'. It is known that the ferro-quadrupole correlation function for this ground state is bounded away from zero. We should note that the bilinear term is ferromagnetic in the region $J' > J > 0$. The strong quantum fluctuation induced by the biquadratic term stabilizes the spin singlet state. In the case of one dimension it is expected that the translation invariance is spontaneously broken in the infinite-volume limit. We think that in two or more dimensions whether QLRO exists or not for $J' > J > 0$ is not at all trivial. It is desirable to confirm the existence of QLRO rigorously.

For the usual d-dimensional spin-S antiferromagnetic Heisenberg model ($J < 0, J' = 0$ in our Hamiltonian (1)), the existence of DLRO can be proved by using the method of infrared bounds. Dyson, Lieb and Simon (DLS) proved that DLRO exists at sufficiently low temperatures if $d \geqslant 3$ and $S \geqslant 1$ [25]. By applying the technique of DLS, Jardão Neves and Fernando Perez proved the existence of DLRO in the ground state for $d = 2$ and $S \geqslant 1$ [26,27]. Kennedy, Lieb and Shastry (KLS) improved the technique of DLS and proved the existence of DLRO in the ground state for $d = 3$ and $S = 1/2$ [28].

In classical cases where $\boldsymbol{S}(x)$ in Hamiltonian (1) is a classical vector, there are some rigorous results on the existence of dipole LRO and quadrupole LRO at finite temperatures. In this case Hamiltonian (1) is related to a model of the nematic phase transitions in liquid crystals. With the method of infrared bounds, Angelescu and Zagrebnov proved the existence of the nematic (ferro-quadrupole) phase transition at low temperatures for a lattice-gas version of the purely nematogenic model ($J = 0, J' > 0$) in three or more dimensions [29] (see also [30] and [31], in which the purely nematogenic lattice-gas models with long-range interactions are discussed). For $J > 0, J' > 0$ Tanaka and Idogaki and Campbell and Chayes proved the existence of ferro-dipole LRO and ferro-quadrupole LRO at low temperatures in three or more dimensions [32, 33]. They also established that there is an intermediate-temperature region where ferro-quadrupole LRO exists but ferro-dipole LRO does not.

The method of infrared bounds can be applied to spin systems satisfying so-called reflection positivity. In this paper we show that Hamiltonian (1) satisfies reflection positivity for $J' \geqslant 0, J' \geqslant 2J$, and following the paper of KLS [28] we prove that, for a restricted parameter region within $J' \geqslant 0, J' \geqslant 2J$, DLRO exists in the ground state of Hamiltonian (1) in two and three dimensions. We further extend the proof to the case of QLRO and examine its existence.

This paper is organized as follows. In section 2, we introduce some notation used throughout this paper. In sections 3 and 4, by using the method of infrared bounds with the KLS technique, we prove the existence of DLRO and QLRO, respectively, in the ground state of Hamiltonian (1). In section 5, we summarize and discuss the results of sections 3 and 4.

2. Definition and notation

We start with the definition of the model. Consider a d-dimensional hypercubic lattice $\Lambda \subset Z^d$ of the form

$$\Lambda = \{x = (x_1, \dots, x_d) | -L+1 \leqslant x_i \leqslant L\} \tag{2}$$

where L is an integer. We impose periodic boundary conditions in all directions. With each site $x \in \Lambda$, we associate the $S = 1$ operator $\boldsymbol{S}(x)$. In this paper we use the usual matrix representation

$$S_1 = \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix} \qquad S_2 = \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & -\mathrm{i} & 0 \\ \mathrm{i} & 0 & -\mathrm{i} \\ 0 & \mathrm{i} & 0 \end{pmatrix} \qquad S_3 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}. \tag{3}$$

We define the isotropic bilinear–biquadratic exchange Hamiltonian as follows:

$$\mathcal{H}_\Lambda = -J \sum_{\langle x,y \rangle} \boldsymbol{S}(x) \cdot \boldsymbol{S}(y) - J' \sum_{\langle x,y \rangle} (\boldsymbol{S}(x) \cdot \boldsymbol{S}(y))^2 \tag{4}$$

where the summation is over all nearest-neighbour pairs $\langle x, y \rangle$ in Λ. The parameters J and J' are restricted to $J' \geqslant 0, J' \geqslant 2J (J \neq 0)$, where we can use the method of infrared bounds in the paper of KLS. The reason for this restriction will become clear in the next section.

For subsequent discussion, it is convenient to rewrite $\mathcal{H}_\Lambda$ in terms of the Racah operators O_{kq}, which are defined by

$$O_{10}(x) = S_3(x) \tag{5}$$

$$O_{1\pm1}(x) = \mp\sqrt{\tfrac{1}{2}}\tfrac{1}{2}\left(S^{\pm}(x) + S^{\pm}(x)\right) \tag{6}$$

$$O_{20}(x) = \tfrac{1}{2}\left[3S_3^2(x) - S(S+1)\right] \tag{7}$$

$$O_{2\pm1}(x) = \mp\sqrt{\tfrac{3}{2}}\tfrac{1}{2}\left(S_3(x)S^{\pm}(x) + S^{\pm}(x)S_3(x)\right) \tag{8}$$

$$O_{2\pm2}(x) = \sqrt{\tfrac{3}{8}}\left(S^{\pm}(x)\right)^2 \tag{9}$$

for $k = 1, 2$ and $|q| \leqslant k$ [34], where $S^+(x)$ and $S^-(x)$ are the usual spin raising and lowering operators defined by

$$S^{\pm}(x) = S_1(x) \pm \mathrm{i}S_2(x). \tag{10}$$

The Racah operators satisfy the relations

$$O^{\dagger}_{kq}(x) = (-1)^q O_{k-q}(x) \tag{11}$$

$$\sum_{q=-1}^{1} O_{1q}(x) O^{\dagger}_{1q}(y) = \boldsymbol{S}(x)\cdot\boldsymbol{S}(y) \tag{12}$$

$$\sum_{q=-2}^{2} O_{2q}(x) O^{\dagger}_{2q}(y) = \tfrac{3}{2}(\boldsymbol{S}(x)\cdot\boldsymbol{S}(y))^2 + \tfrac{3}{4}\boldsymbol{S}(x)\cdot\boldsymbol{S}(y) - \tfrac{1}{2}S^2(S+1)^2 \tag{13}$$

and the commutation relation,

$$\left[S_3(x), O_{kq}(x)\right] = q\, O_{kq}(x). \tag{14}$$

By using equations (12) and (13), Hamiltonian (4) can be written as

$$\mathcal{H}_\Lambda = J_\mathrm{D}\mathcal{H}_{\Lambda\mathrm{D}} - J_\mathrm{Q}\mathcal{H}_{\Lambda\mathrm{Q}} \tag{15}$$

with

$$J_\mathrm{D} = -J + \tfrac{1}{2}J' \qquad J_\mathrm{Q} = \tfrac{2}{3}J' \tag{16}$$

$$\mathcal{H}_{\Lambda\mathrm{D}} = \sum_{\langle x,y\rangle}\sum_{q=-1}^{1} O_{1q}(x)O^{\dagger}_{1q}(y) \tag{17}$$

$$\mathcal{H}_{\Lambda\mathrm{Q}} = \sum_{\langle x,y\rangle}\sum_{q=-2}^{2} O_{2q}(x)O^{\dagger}_{2q}(y) \tag{18}$$

where we have omitted a constant term. From $J' \geqslant 0$, $J' \geqslant 2J$, the parameters J_D and J_Q are restricted to $J_\mathrm{D} \geqslant 0$, $J_\mathrm{Q} \geqslant 0$.

We define a DLRO parameter and a QLRO parameter in the ground state of $\mathcal{H}_\Lambda$ by

$$m_\mathrm{D}^2 = \lim_{\Lambda\to\infty}\left\langle\left[\frac{1}{|\Lambda|}\sum_{x\in\Lambda}(-1)^x S_3(x)\right]^2\right\rangle_\Lambda \tag{19}$$

and

$$m_\mathrm{Q}^2 = \lim_{\Lambda\to\infty}\left\langle\left(\frac{1}{|\Lambda|}\sum_{x\in\Lambda}O_{20}(x)\right)^2\right\rangle_\Lambda \tag{20}$$

respectively, where $|\Lambda|$ is the number of sites in Λ and $\langle X\rangle_\Lambda$ denotes an expectation value of X for the ground state.

3. Dipole long-range order

In this section, we prove the existence of DLRO, i.e. $m_\mathrm{D}^2 > 0$ within the region $J < 0$ and $J' \geqslant 0$ ($0 \leqslant J_\mathrm{Q} < 4J_\mathrm{D}/3$).

Let us define the Fourier transform of $S_3(x)$ by

$$S_3(k) = \frac{1}{\sqrt{|\Lambda|}}\sum_{x\in\Lambda}\mathrm{e}^{-\mathrm{i}k\cdot x}S_3(x) \tag{21}$$

where $k = (k_1, \ldots, k_d)$ is in a reciprocal lattice Λ^*. The Fourier transform of the two-point correlation function in the ground state is written as

$$g_\mathrm{D}(k) = \langle S_3(k)S_3(-k)\rangle_\Lambda. \tag{22}$$

Let us define the dipole interaction energy and the quadrupole interaction energy per site by

$$e_{\Lambda D} = -\frac{\langle \mathcal{H}_{\Lambda D} \rangle_\Lambda}{|\Lambda|} \qquad e_D = \lim_{\Lambda \to \infty} e_{\Lambda D} \tag{23}$$

and

$$e_{\Lambda Q} = \frac{\langle \mathcal{H}_{\Lambda Q} \rangle_\Lambda}{|\Lambda|} \qquad e_Q = \lim_{\Lambda \to \infty} e_{\Lambda Q} \tag{24}$$

respectively. By using the sum rule

$$\frac{1}{|\Lambda|} \sum_{k \in \Lambda^*} g_D(k) \frac{1}{d} \sum_{m=1}^{d} \cos k_m = -\frac{e_{\Lambda D}}{3d} \tag{25}$$

the DLRO parameter can be written as

$$m_D^2 = \lim_{\Lambda \to \infty} \frac{1}{|\Lambda|} g_D(Q) = \frac{e_D}{3d} - G_D \qquad Q = (\pi, \ldots, \pi) \tag{26}$$

with

$$G_D = \lim_{\Lambda \to \infty} \frac{1}{|\Lambda|} \sum_{k \neq Q} g_D(k) \left(-\frac{1}{d} \sum_{m=1}^{d} \cos k_m \right). \tag{27}$$

In order to estimate a lower bound for the right-hand side of equation (26), first, let us consider a lower bound on e_D. Taking the Néel state as a variational state for Hamiltonian (15), we obtain

$$-J_D e_D - J_Q e_Q \leqslant -J_D S^2 d - \tfrac{1}{4} J_Q S^2 (2S-1)^2 d = -J_D d - \tfrac{1}{4} J_Q d. \tag{28}$$

As we shall see in the next section (see inequality (80)), we have

$$e_D \geqslant \tfrac{4}{5} e_Q \tag{29}$$

for $J < 0$, and therefore we obtain

$$e_D \geqslant d\left(1 + \tfrac{1}{4}\alpha\right)\left(1 + \tfrac{5}{4}\alpha\right)^{-1} \tag{30}$$

with $\alpha = J_Q / J_D$.

Next we estimate an upper bound on G_D. As in the KLS method [28], we introduce the spectral weight function

$$R_D(\omega) = \tfrac{1}{2} \sum_n \left(|\langle \psi_n | S_3(k) | \psi_0 \rangle|^2 + |\langle \psi_n | S_3(-k) | \psi_0 \rangle|^2\right) \delta(\omega - e_n + e_0) \tag{31}$$

where ψ_n are the energy eigenstates, e_n are the corresponding eigenvalues and ψ_0 is the nondegenerate ground state of Hamiltonian (15). Then,

$$g_D(k) = \int_0^\infty \mathrm{d}\omega \, R_D(\omega). \tag{32}$$

By using the Cauchy–Schwarz inequality, we have

$$(g_D(k))^2 = \left(\int_0^\infty \mathrm{d}\omega \, R_D(\omega) \right)^2 \leqslant \int_0^\infty \mathrm{d}\omega \, R_D(\omega) \omega^{-1} \int_0^\infty \mathrm{d}\omega \, R_D(\omega) \omega. \tag{33}$$

Noting the relation

$$\langle O_{2q}(x) O_{2q}^\dagger(y) \rangle_\Lambda = \frac{e_{\Lambda Q}}{5d} \tag{34}$$

the last integral of the right-hand side of inequality (33) is reduced to

$$\int_0^\infty \mathrm{d}\omega \, R_D(\omega) \omega = \frac{1}{2} \langle [[S_3(k), \mathcal{H}_\Lambda], S_3(-k)] \rangle_\Lambda = \frac{1}{d} \left(\frac{2}{3} J_D e_{\Lambda D} + 2 J_Q e_{\Lambda Q} \right) E(k) \tag{35}$$

with

$$E(k) = \sum_{m=1}^{d} (1 - \cos k_m). \tag{36}$$

We shall show later that

$$\int_0^\infty \mathrm{d}\omega\, R_\mathrm{D}(\omega)\omega^{-1} \leqslant \frac{1}{4J_\mathrm{D}E(k-Q)} \qquad k \neq Q. \tag{37}$$

Therefore, from equation (35) and inequalities (33) and (37), we obtain

$$g_\mathrm{D}(k) \leqslant \sqrt{\frac{E(k)}{E(k-Q)}}\sqrt{\frac{e_{\Lambda\mathrm{D}}}{6d} + \frac{e_{\Lambda\mathrm{Q}}}{2d}\alpha}. \tag{38}$$

By using inequalities (29) and (38), G_D is bounded from above by

$$\overline{G}_\mathrm{D} = \sqrt{\frac{e_\mathrm{D}}{d}\left(\frac{1}{6} + \frac{5}{8}\alpha\right)}\Gamma_\mathrm{I}(d) \tag{39}$$

with

$$\Gamma_\mathrm{I}(d) = \frac{1}{(2\pi)^d}\int_{-\pi}^{\pi} \mathrm{d}^d k \sqrt{\frac{E(k)}{E(k-Q)}}\left\{-\frac{1}{d}\sum_{m=1}^{d}\cos k_m\right\}_+ \tag{40}$$

where

$$\{F\}_+ = \begin{cases} F & \text{if} \quad F \geqslant 0 \\ 0 & \text{otherwise.} \end{cases} \tag{41}$$

We evaluate numerically

$$\begin{aligned} \Gamma_\mathrm{I}(2) &= 0.646\ldots \\ \Gamma_\mathrm{I}(3) &= 0.349\ldots. \end{aligned} \tag{42}$$

From equations (26) and (39) and inequality (30), we obtain

$$m_\mathrm{D}^2 \geqslant \sqrt{\frac{e_\mathrm{D}}{d}}\left(\frac{1}{3}\sqrt{\frac{e_\mathrm{D}}{d}} - \sqrt{\left(\frac{1}{6} + \frac{5}{8}\alpha\right)}\Gamma_\mathrm{I}(d)\right) \tag{43}$$

$$\geqslant \sqrt{\frac{e_\mathrm{D}}{d}}\left(\frac{1}{3}\sqrt{\left(1 + \frac{1}{4}\alpha\right)\left(1 + \frac{5}{4}\alpha\right)^{-1}} - \sqrt{\left(\frac{1}{6} + \frac{5}{8}\alpha\right)}\Gamma_\mathrm{I}(d)\right). \tag{44}$$

Therefore the DLRO parameter takes a strictly positive value if the parameter α satisifies

$$\alpha < 0.115 \qquad d = 2 \tag{45}$$

$$\alpha < 0.659 \qquad d = 3 \tag{46}$$

or if the parameters J and J' satisfy

$$J' < -0.188J \qquad d = 2 \tag{47}$$

$$J' < -1.954J \qquad d = 3. \tag{48}$$

In the following we shall prove inequality (37). Let us define

$$T_1(x) = U^\dagger S_1(x) U \tag{49}$$

$$T_2(x) = U^\dagger \mathrm{i} S_2(x) U \tag{50}$$

$$T_3(x) = U^\dagger S_3(x) U \tag{51}$$

with $U = \exp(\mathrm{i}\pi \sum_{x\in\Lambda_{\rm odd}} S_2(x))$, where $\Lambda_{\rm odd}$ is a collection of sites x with odd $|x|$. (For $x = (x_1, ..., x_d)$ $|x| = \sum_{i=1}^d |x_i|$.) Let $h(x)$ be a real-valued function on the sites. Then we consider the h-dependent Hamiltonian defined by

$$\begin{aligned}\mathcal{H}_\Lambda^{\rm D}(h) = -J_{\rm D} \sum_{\langle x,y\rangle} \{&T_1(x)T_1(y) + T_2(x)T_2(y) + (T_3(x) - h(x))(T_3(y) - h(y)) \\ &+\tfrac{1}{2}[T_3^2(x) - (T_3(x) - h(x))^2 + T_3^2(y) - (T_3(y) - h(y))^2]\} \\ &-J_{\rm Q} \sum_{\langle x,y\rangle} \sum_{q=-2}^{2} O_{2q}(x)O_{2q}(y). \end{aligned} \tag{52}$$

We note that

$$\mathcal{H}_\Lambda^{\rm D}(0) = U^\dagger \mathcal{H}_\Lambda U. \tag{53}$$

For $\mathcal{H}_\Lambda^{\rm D}(h)$ we can easily check the following facts: the matrix elements of all the matrices appearing in $\mathcal{H}_\Lambda^{\rm D}(h)$ are real, the coefficients of all the nearest-neighbour dipole–dipole interaction terms are negative for $J_{\rm D} > 0$ and those of nearest-neighbour quadrupole–quadrupole interaction terms are non-positive for $J_{\rm Q} \geqslant 0$. These conditions are required so as to satisfy reflection positivity. Thus, following the argument of KLS (p 1026 in [28]), we have

$$E_\Lambda^{\rm D}(h) \geqslant E_\Lambda^{\rm D}(0) \tag{54}$$

which leads to

$$\frac{\mathrm{d}^2}{\mathrm{d}\lambda^2} E_\Lambda^{\rm D}(\lambda h)\Big|_{\lambda=0} \geqslant 0 \tag{55}$$

where $E_\Lambda^{\rm D}(h)$ denotes the ground-state energy of $\mathcal{H}_\Lambda^{\rm D}(h)$. Using inequality (55) and a second-order perturbation theory for $\mathcal{H}_\Lambda^{\rm D}(\lambda h)$ with $h(x) = \cos k\cdot x$ and $\sin k \cdot x$, we obtain inequality (37).

4. Quadrupole long-range order

In this section, we prove the existence of QLRO within the region $J' \geqslant 2J > 0$ $(0 \leqslant 4J_{\rm D}/3 < J_{\rm Q})$. As in section 3, let us define the Fourier transform of $O_{20}(x)$,

$$O_{20}(k) = \frac{1}{\sqrt{|\Lambda|}} \sum_{x\in\Lambda} \mathrm{e}^{-\mathrm{i}k\cdot x} O_{20}(x). \tag{56}$$

The Fourier-transformed two-point quadrupole correlation function in the ground state is

$$g_{\rm Q}(k) = \langle O_{20}(k) O_{20}(-k)\rangle_\Lambda. \tag{57}$$

By using $g_{\rm Q}(k)$ and the sum rule

$$\frac{1}{|\Lambda|} \sum_{k\in\Lambda^*} g_{\rm Q}(k) \frac{1}{d} \sum_{m=1}^{d} \cos k_m = \frac{e_{\Lambda\rm Q}}{5d} \tag{58}$$

the QLRO parameter is written as

$$m_{\rm Q}^2 = \lim_{\Lambda\to\infty} \frac{1}{|\Lambda|} g_{\rm Q}(0) = \frac{e_{\rm Q}}{5d} - G_{\rm Q} \qquad 0 = (0, \ldots, 0) \tag{59}$$

with

$$G_{\rm Q} = \lim_{\Lambda\to\infty} \frac{1}{|\Lambda|} \sum_{k\neq 0} g_{\rm Q}(k) \frac{1}{d} \sum_{m=1}^{d} \cos k_m. \tag{60}$$

To evaluate a lower bound for the right-hand side of equation (59), we follow a similar process in section 3. Firstly, we use the variational method to estimate a lower bound for e_{Q}. Let $|0\rangle_x$ be the eigenstate of $S_3(x)$ with the eigenvalue zero. If we take $\otimes_{x\in\Lambda}|0\rangle_x$ as a trial state for Hamiltonian (15), we have

$$-J_{\mathrm{D}}e_{\mathrm{D}} - J_{\mathrm{Q}}e_{\mathrm{Q}} \leqslant -J_{\mathrm{Q}}d. \tag{61}$$

We shall see later that

$$e_{\mathrm{D}} \leqslant \tfrac{4}{5}e_{\mathrm{Q}} \tag{62}$$

for $J > 0$ (see inequality (79)), and thus we obtain

$$e_{\mathrm{Q}} \geqslant d(1 + \tfrac{4}{5}\beta)^{-1} \tag{63}$$

with $\beta = J_{\mathrm{D}}/J_{\mathrm{Q}}$ ($\beta = 1/\alpha$).

Secondly, we estimate an upper bound on G_{Q}. We use the spectral weight function

$$R_{\mathrm{Q}}(\omega) = \tfrac{1}{2}\sum_n \left(\left|\langle\psi_n|O_{20}(k)|\psi_0\rangle\right|^2 + \left|\langle\psi_n|O_{20}(-k)|\psi_0\rangle\right|^2\right)\delta(\omega - e_n + e_0). \tag{64}$$

By using the Cauchy–Schwarz inequality,

$$(g_{\mathrm{Q}}(k))^2 = \left(\int_0^\infty \mathrm{d}\omega\, R_{\mathrm{Q}}(\omega)\right)^2 \leqslant \int_0^\infty \mathrm{d}\omega\, R_{\mathrm{Q}}(\omega)\omega^{-1} \int_0^\infty \mathrm{d}\omega\, R_{\mathrm{Q}}(\omega)\omega. \tag{65}$$

The last integral of the right-hand side of equation (65) takes the form

$$\begin{aligned}\int_0^\infty \mathrm{d}\omega\, R_{\mathrm{Q}}(\omega)\omega &= \tfrac{1}{2}\big\langle\big[\big[O_{20}(k), \mathcal{H}_\Lambda\big], O_{20}(-k)\big]\big\rangle_\Lambda \\ &= J_{\mathrm{D}}\left(\frac{6}{5d}e_{\Lambda\mathrm{Q}}\sum_m^d \cos k_m + \frac{3}{2}e_{\Lambda\mathrm{D}}\right) + J_{\mathrm{Q}}\left(\frac{9}{8d}e_{\Lambda\mathrm{D}}\sum_m^d \cos k_m + \frac{9}{10}e_{\Lambda\mathrm{Q}}\right)\end{aligned} \tag{66}$$

where we have used the following commutation relations for $S = 1$:

$$\big[O_{20}(x), O_{1\pm1}(x)\big] = \pm\sqrt{3}O_{2\pm1}(x) \tag{67}$$

$$\big[O_{20}(x), O_{2\pm1}(x)\big] = \pm\frac{3\sqrt{3}}{4}O_{1\pm1}(x) \tag{68}$$

$$\big[O_{20}(x), O_{2\pm2}(x)\big] = 0. \tag{69}$$

We note that these commutation relations hold only in the case of $S = 1$. An upper bound on the first integral of the right-hand side of equation (65) can be derived as

$$\int_0^\infty \mathrm{d}\omega\, R_{\mathrm{Q}}(\omega)\omega^{-1} \leqslant \frac{1}{4J_{\mathrm{Q}}E(k)} \qquad k \neq 0. \tag{70}$$

To prove inequality (70), we take the h-dependent unitary transformed Hamiltonian,

$$\begin{aligned}\mathcal{H}_\Lambda^{\mathrm{Q}}(h) = &-J_{\mathrm{D}}\sum_{\langle x,y\rangle} \boldsymbol{T}(x)\cdot\boldsymbol{T}(y) \\ &-J_{\mathrm{Q}}\sum_{\langle x,y\rangle}\Bigg\{\sum_{q\neq0} O_{2q}(x)O_{2q}(y) + (O_{20}(x) - h(x))(O_{20}(y) - h(y)) \\ &+\tfrac{1}{2}\big[O_{20}^2(x) - (O_{20}(x) - h(x))^2 + O_{20}^2(y) - (O_{20}(y) - h(y))^2\big]\Bigg\}.\end{aligned} \tag{71}$$

As in the case of $\mathcal{H}_\Lambda^{\mathrm{D}}(h)$, we can easily check that the Hamiltonian $\mathcal{H}_\Lambda^{\mathrm{Q}}(h)$ satisfies reflection positivity for $J' \geqslant 2J$, $J' > 0$. Thus, following the argument of KLS and using the second-order perturbation theory, we can obtain inequality (70).

Here we note that inequality (70) holds for $k \neq 0$, while inequality (37) holds for $k \neq Q$. This is roughly explained as follows. First we note that $\mathcal{H}_\Lambda^{\mathrm{D}}(0) = \mathcal{H}_\Lambda^{\mathrm{Q}}(0) = U^\dagger \mathcal{H}_\Lambda U$ and that the eigenstates $\tilde{\psi}_n$ of $\mathcal{H}_\Lambda^{\mathrm{D}}(0)$ and $\mathcal{H}_\Lambda^{\mathrm{Q}}(0)$ are related to the eigenstates ψ_n of $\mathcal{H}_\Lambda$ by $\tilde{\psi}_n = U^\dagger \psi_n$. In the second-order perturbation theory we need to calculate

$$\left\langle \tilde{\psi}_n \left| \sum_{\langle x,y \rangle} (T_3(x) - T_3(y))(h(x) - h(y)) \right| \tilde{\psi}_0 \right\rangle \tag{72}$$

in the DLRO case and

$$\left\langle \tilde{\psi}_n \left| \sum_{\langle x,y \rangle} (O_{20}(x) - O_{20}(y))(h(x) - h(y)) \right| \tilde{\psi}_0 \right\rangle \tag{73}$$

in the QLRO case. Expression (72) is reduced to

$$\left\langle \psi_n \left| \sum_{\langle x,y \rangle} (\mathrm{e}^{\mathrm{i}Qx} S_3(x) - \mathrm{e}^{\mathrm{i}Qx} S_3(y))(h(x) - h(y)) \right| \psi_0 \right\rangle \tag{74}$$

and then the factor $\mathrm{e}^{\mathrm{i}Qx}$ appears; this extra factor leads to the phase shift by Q in inequality (37). On the other hand, since $U O_{20}(x) U^\dagger = O_{20}(x)$, no extra factor appears in equation (73) and thus inequality (70) holds for $k \neq 0$.

We turn back to an estimate of an upper bound on G_{Q}. From equation (66) and inequalities (65) and (70) we obtain

$$g_{\mathrm{Q}}(k) \leqslant \sqrt{\frac{I_{\Lambda 1}\left\{\frac{1}{d}\sum_{m=1}^{d}\cos k_m\right\} + I_{\Lambda 2}}{E(k)}} \tag{75}$$

with

$$I_{\Lambda 1} = \tfrac{9}{32} e_{\Lambda \mathrm{D}} + \tfrac{3}{10} \beta e_{\Lambda \mathrm{Q}} \tag{76}$$

$$I_{\Lambda 2} = \tfrac{3}{8} \beta e_{\Lambda \mathrm{D}} + \tfrac{9}{40} e_{\Lambda \mathrm{Q}}. \tag{77}$$

Then we have

$$\frac{1}{|\Lambda|}\sum_{k\neq 0} g_{\mathrm{Q}}(k)\frac{1}{d}\sum_{m=1}^{d}\cos k_m \leqslant \frac{1}{|\Lambda|}\sum_{k\neq 0}\sqrt{\frac{I_{\Lambda 1}\left\{\frac{1}{d}\sum_{m=1}^{d}\cos k_m\right\}_+ + I_{\Lambda 2}}{E(k)}}\left\{\frac{1}{d}\sum_{m=1}^{d}\cos k_m\right\}_+ . \tag{78}$$

Noting that $R_{\mathrm{Q}}(\omega)$ is non-negative, we find that equation (66) is also non-negative. Thus, the expectation value of double commutator (66) at $k = Q$ leads to

$$e_{\Lambda \mathrm{D}} \leqslant \tfrac{4}{5} e_{\Lambda \mathrm{Q}} \qquad J > 0 \tag{79}$$

$$e_{\Lambda \mathrm{D}} \geqslant \tfrac{4}{5} e_{\Lambda \mathrm{Q}} \qquad J < 0. \tag{80}$$

Applying inequality (79) to equations (76) and (77), we have

$$I_{\Lambda 1} \leqslant \left(\tfrac{9}{40} + \tfrac{3}{10}\beta\right) e_{\Lambda \mathrm{Q}} \tag{81}$$

$$I_{\Lambda 2} \leqslant \left(\tfrac{9}{40} + \tfrac{3}{10}\beta\right) e_{\Lambda \mathrm{Q}} \tag{82}$$

for $J > 0$.

From inequalities (78), (81) and (82), G_{Q} is bounded from above by $\overline{G}_{\mathrm{Q}}$ with

$$\overline{G}_{\mathrm{Q}} = \sqrt{\left(\tfrac{9}{40} + \tfrac{3}{10}\beta\right) e_{\mathrm{Q}}}\,\Gamma_{\mathrm{II}}(d) \tag{83}$$

where

$$\Gamma_{\mathrm{II}}(d) = \frac{1}{(2\pi)^d}\int_{-\pi}^{\pi} \mathrm{d}^d k \sqrt{\frac{\left\{\frac{1}{d}\sum_{m=1}^{d}\cos k_m\right\}_+ + 1}{E(k)}} \left\{\frac{1}{d}\sum_{m=1}^{d}\cos k_m\right\}_+ . \qquad (84)$$

We calculate numerically

$$\begin{aligned}\Gamma_{\mathrm{II}}(2) &= 0.457\ldots \\ \Gamma_{\mathrm{II}}(3) &= 0.202\ldots.\end{aligned} \qquad (85)$$

From equations (59) and (83) and inequality (63), we obtain

$$m_{\mathrm{Q}}^2 \geqslant \sqrt{e_{\mathrm{Q}}}\left(\frac{1}{5d}\sqrt{e_{\mathrm{Q}}} - \sqrt{\frac{9}{40} + \frac{3}{10}\beta\Gamma_{\mathrm{II}}(d)}\right) \qquad (86)$$

$$\geqslant \sqrt{e_{\mathrm{Q}}}\left(\frac{1}{5d}\sqrt{d\left(1+\frac{4}{5}\beta\right)^{-1}} - \sqrt{\frac{9}{40} + \frac{3}{10}\beta\Gamma_{\mathrm{II}}(d)}\right). \qquad (87)$$

Consequently, in three dimensions the QLRO parameter takes a non-zero value for

$$\beta < 0.187 \qquad (88)$$

or

$$J' < 2.664J. \qquad (89)$$

In two dimensions there is no region for which we can prove $m_{\mathrm{Q}}^2 > 0$ from the lower bound (87).

5. Summary and discussions

We have proved that the DLRO or the QLRO parameter takes a finite value under some conditions. The results are summarized in figure 1. The boundaries $J' = 2J, J > 0$ and $J' = 0, J < 0$ in figure 1 originate from the condition for reflection positivity. Thus it is impossible to improve these boundaries within the present method. On the other hand, we have a chance to improve the other boundaries by estimating better lower bounds on e_{D} and e_{Q} or better upper bounds on G_{D} and G_{Q}, which lead to improved lower bounds on m_{D}^2 and m_{Q}^2. One candidate for doing so is to find more suitable trial states, which give us lower variational energies, by which we can obtain bounds on e_{D} and e_{Q}.

It should be noted, however, that in the case of two dimensions we cannot conclude from inequality (86) the existence of QLRO, even though we find the exact value of e_{Q}. The reason is as follows. Suppose that the right-hand side of (86) is positive. Then, we have $e_{\mathrm{Q}} > 4.699$. On the other hand, we have found $e_{\mathrm{Q}} < 2.578$ by the numerical calculation (see appendix A). This implies the fact that the right-hand side of (86) never takes a positive value.

Within the region $J' > J > 0$, which is thought to be the FQ phase by approximation methods [14–16, 18–20], we have rigorously established the existence of QLRO in three dimensions. For the parameter regions where we have proved the existence of QLRO, following the argument by Kaplan, Horsch and von der Linden [35] (see also [36] and [37]), we can construct an infinite-volume ground state with explicit symmetry breaking by applying an infinitesimal symmetry breaking field. Thus our results partially confirm predictions by approximation methods. In order to show the occurrence of the FQ phase, furthermore, we need to prove that the expectation value of the dipole moment is vanishing for any infinite-volume ground states. The recent numerical results of Harada and Kawashima by means of a quantum Monte Carlo method indicate that there is no dipole moment in the ground state

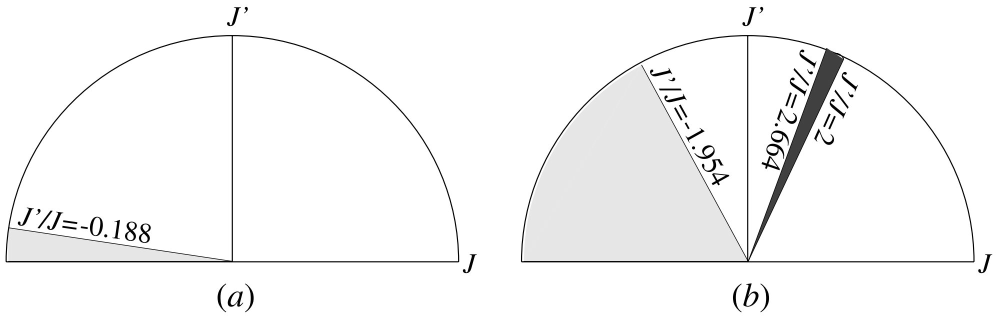

Figure 1. The region in which we have established the existence of LRO. (*a*) The square lattice. (*b*) The simple cubic lattice. The DLRO and the QLRO parameters take a non-zero value in the light-grey and the dark-grey regions, respectively.

for $J' > J > 0$ in two dimensions [21]. They showed that the algorithm for $J' > J > 0$ is different from that for $0 \leqslant J' < J$ where the system is ferromagnetic and also different from that for $J < 0$, $J' \geqslant 0$ where the system is expected to be antiferromagnetic. They pointed out that the algorithmic 'transition' points $J = 0$ and $J' = J$ can be related to the phase transition points in real physics. They also pointed out that the algorithmic 'transition' points do not depend on the lattice dimensionality. Therefore we expect the absence of dipole moment for $J' > J > 0$ in three dimensions. However, the rigorous proof of this is far beyond our scope, and we leave it as an interesting future problem to be performed.

In the following we shall comment on the case $S > 1$. In this case, we should consider Hamiltonian (4) with J and J' replaced by J/S^2 and J'/S^4. By using the variational argument [13], we easily find that the ground state is ferromagnetic for $0 < J' < SJ$. Thus, as S increases, the ferromagnetic region becomes larger and the FQ phase (if it exists) becomes smaller in the J–J' plane. It should be noted that there is no quadrupole region in the ground state for $J > 0$ in the limit $S \to \infty$, and that its existence is caused by quantum effects. We can also easily check that the system satisfies reflection positivity for $J' \geqslant 2S^2 J$, $J' \geqslant 0$. Thus, the present method is applicable to such a parameter region, provided that the finite-volume ground state is unique. It is proved that the finite-volume ground state in the region $J' < -SJ/2(S-1)$, $J' \geqslant 0$ is unique [22], and as far as we know there is no rigorous proof of it in the other region. Under the assumption that the finite-volume ground state in the region satisfying the reflection positivity is unique, we can obtain the same inequalities as (37) and (70). Then, following the same method as in section 3, we can prove the existence of DLRO (see appendix B). On the other hand, in the case of QLRO, difficulties will arise in calculating an upper bound on the second integral of equation (65); unlike the $S = 1$ case in which Racah operators satisfy the commutation relations (67)–(69), an expression of the expectation value of double commutator (66) takes a complicated form for $S > 1$. Thus it is not easy to prove the existence of QLRO in the case of $S > 1$ by the present method.

Acknowledgments

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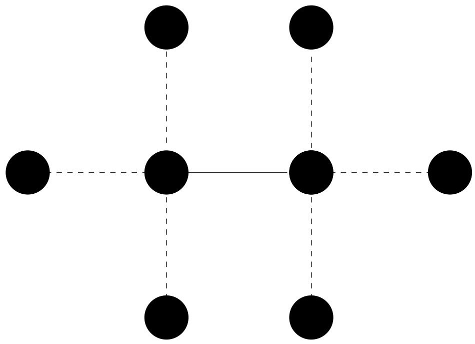

Figure A.1. The eight-site cluster used to derive a bounds on $e_{\Lambda Q}$ for the square lattice. The solid and dotted lines represent full- and half-weight bonds.

Appendix A

In this appendix we numerically estimate an upper bound on $e_{\Lambda Q}$, relying on the computer.

Let Λ' be a subset of Λ including the origin, and define a local Hamiltonian by

$$h_Q^n(0) = \sum_{x\in\Lambda'} h_Q(x) \tag{90}$$

where $|\Lambda'| = n$ and

$$h_Q(x) = \frac{1}{2}\sum_{m=1}^{d}\sum_{q=-2}^{2} O_{2q}(x)\big(O_{2q}^\dagger(x+\delta_m) + O_{2q}^\dagger(x-\delta_m)\big) \tag{91}$$

where δ_m denotes the unit vector in the m-direction. Then, the whole Hamiltonian is written as

$$\mathcal{H}_{\Lambda Q} = \frac{1}{n}\sum_{x\in\Lambda} h_Q^n(x) \tag{92}$$

where $h_Q^n(x)$ is a translated copy of $h_Q^n(0)$. Thus, by calculating the maximum eigenvalue $\bar{e}_Q^n$ of $h_Q^n(x)$, we can obtain an inequality

$$e_{\Lambda Q} \leqslant \frac{1}{n}\bar{e}_Q^n. \tag{93}$$

Turning now to $d = 2$, we adopt $h_Q^2(0)$, whose support is described in figure A.1. By using the Householder method, we have calculated all the eigenvalues of $h_Q^2(0)$. The result is $\bar{e}_Q^2 = 5.154\ldots$, which gives us

$$e_{\Lambda Q} \leqslant 2.578. \tag{94}$$

Appendix B

In this appendix we are concerned with the proof of the existence of DLRO in the ground state of the Hamiltonian

$$\mathcal{H} = -\frac{J}{S^2}\sum_{\langle x,y\rangle} \boldsymbol{S}(x)\cdot\boldsymbol{S}(y) - \frac{J'}{S^4}\sum_{\langle x,y\rangle}(\boldsymbol{S}(x)\cdot\boldsymbol{S}(y))^2 \tag{95}$$

with $S > 1$. By using equations (17) and (18), Hamiltonian (95) is written as

$$\mathcal{H}_\Lambda = \widetilde{J}_\mathrm{D}\mathcal{H}_{\Lambda\mathrm{D}} - \widetilde{J}_\mathrm{Q}\mathcal{H}_{\Lambda\mathrm{Q}} \tag{96}$$

with

$$\widetilde{J}_\mathrm{D} = -\frac{J}{S^2} + \frac{1}{2}\frac{J'}{S^4} \qquad \widetilde{J}_\mathrm{Q} = \frac{2}{3}\frac{J'}{S^4} \tag{97}$$

where we have omitted a constant term.

We can easily see that Hamiltonian (96) satisfies reflection positivity for $\widetilde{J}_\mathrm{D} \geqslant 0$, $\widetilde{J}_\mathrm{Q} \geqslant 0$ or $J \geqslant 0$, $J' \geqslant 2S^2 J$. We assume that the finite-volume ground state of Hamiltonian (95) in this region is unique. Under this assumption, we obtain inequality (38) with α replaced by $\widetilde{\alpha} = \widetilde{J}_\mathrm{Q}/\widetilde{J}_\mathrm{D}$.

The main difference between the cases of $S = 1$ and $S > 1$ arises from the fact that the commutation relations (67)–(69) do not hold for $S > 1$; this implies that the last equality of (66) does not hold, and therefore we cannot obtain inequality (29) for $S > 1$. Thus in order to bound e_D and G_D we need to obtain another bound on e_Q instead of inequality (29) in the case of $S > 1$. Here we use the Cauchy–Schwarz inequality, and then we obtain

$$e_{\Lambda\mathrm{Q}} \leqslant \left\{S^2(S+1)^2 - \tfrac{3}{4}S(S+1)\right\}d. \tag{98}$$

By using inequality (98), (39) is replaced by

$$\overline{G}_\mathrm{D} = \sqrt{\frac{1}{6d}e_\mathrm{D} + \frac{\widetilde{\alpha}}{2}\left\{S^2(S+1)^2 - \tfrac{3}{4}S(S+1)\right\}}\,\Gamma_\mathrm{I}(d) \tag{99}$$

and, therefore, a lower bound for the DLRO parameter is evaluated as

$$m_\mathrm{D}^2 \geqslant \frac{e_\mathrm{D}}{3d} - \sqrt{\frac{1}{6d}e_\mathrm{D} + \frac{\widetilde{\alpha}}{2}\left\{S^2(S+1)^2 - \tfrac{3}{4}S(S+1)\right\}}\,\Gamma_\mathrm{I}(d). \tag{100}$$

Thus we can conclude $m_\mathrm{D}^2 > 0$ if e_D satisfies

$$e_\mathrm{D} - \left(a + \sqrt{a^2 + \widetilde{\alpha}b}\right) > 0 \tag{101}$$

where

$$a = \tfrac{3}{4}d\Gamma_\mathrm{I}^2(d) \tag{102}$$

$$b = \tfrac{9}{2}d^2\left\{S^2(S+1)^2 - \tfrac{3}{4}S(S+1)\right\}\Gamma_\mathrm{I}^2(d). \tag{103}$$

Let us estimate a lower bound for e_D. Combining inequalities (98) and (28) with J_D and J_Q replaced by $\widetilde{J}_\mathrm{D}$ and $\widetilde{J}_\mathrm{Q}$, we have

$$e_\mathrm{D} \geqslant S^2 d + \widetilde{\alpha}\left(-3S^3 + \tfrac{3}{4}S\right)d. \tag{104}$$

Thus the left-hand side of inequality (101) is bounded from below as

$$e_\mathrm{D} - \left(a + \sqrt{a^2 + \widetilde{\alpha}b}\right) \geqslant S^2 d + \widetilde{\alpha}\left(-3S^3 + \tfrac{3}{4}S\right)d - \left(a + \sqrt{a^2 + \widetilde{\alpha}b}\right). \tag{105}$$

Therefore we obtain the region for which we can prove $m_\mathrm{D}^2 > 0$ as follows:

$$0 \leqslant J' < -\frac{6c}{4-3c}S^2 J \tag{106}$$

with

$$c = \frac{4S^2(2S+1) + 3\Gamma_\mathrm{I}^2(d)(2S^2+3S+2)}{3S(2S-1)(2S+1)^2} - \frac{\sqrt{9\Gamma_\mathrm{I}^4(d)(2S^2+3S+2)^2 + 24S^2\Gamma_\mathrm{I}^2(d)(4S^3+12S^2+11S+3)}}{3S(2S-1)(2S+1)^2}. \tag{107}$$

The results for some values of S are listed in table B.1.

Table B.1. The region in which we have proved the existence of DLRO within the region $J < 0$ and $J' \geqslant 0$.

S	Square lattice	Simple cubic lattice
3/2	$J' < -0.259J$	$J' < -0.514J$
2	$J' < -0.452J$	$J' < -0.731J$
5/2	$J' < -0.650J$	$J' < -0.952J$
3	$J' < -0.853J$	$J' < -1.176J$
∞	$J' < \infty$	$J' < \infty$

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