Solvable models of spin-$\frac{1}{2}$ chains with an energy gap

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(Received 21 March 1994; revised manuscript received 28 August 1995)

An XY model with two kinds of coupling constants J and $-K$ is investigated. The system has plural spin sites in a unit cell, where the coupling constant inside a unit cell is $-K$, and the coupling constant between two unit cells is J. When the number of the spin sites L in a unit cell is even (or odd), there is an energy gap (or no gap) between the ground state and the first excited state (for $|K/J| \neq 1$). The magnetization of the system with any even number of L vanishes below a critical magnetic field at the temperature $T=0$. Therefore, the susceptibility is zero at $T=0$ when L is even, but the susceptibility is positive when L is odd. The data for the magnetization of $(CH_3)_4N$ Ni $(NO_2)_3$ resemble the calculated value of the magnetization for $L=2$.

PACS number(s): 05.50.+q, 75.10.Jm, 75.40.−s

I. INTRODUCTION

Since Haldane [1] conjectured that a one-dimensional Heisenberg antiferromagnet (1DHA) with an integer spin value has an energy gap between the ground state and the first excited state, a number of theoretical studies have been done to elucidate the nature of the Haldane gap. Haldane's conjecture has been confirmed by several numerical calculations [2]. On the other hand, there is no exact analytical treatment of the 1DHA with an integer spin, except the treatment by Affleck *et al.* [3]. They studied the spin $S=1$ 1DHA system with the biquadratic exchange interaction term, and showed the existence of the energy gap exactly. However, there has been no real system close to this model. Thus it is highly desirable to find a realistic Hamiltonian that can be solved exactly. An XY model is a good candidate for this, because the $S=\frac{1}{2}$ one-dimensional XY model has been solved exactly [4] and there are a few examples of real materials which are well approximated by an XY model. The energy spectrum of the traditional XY model is a continuous function of the wave number, and has no energy gap. In this paper, we will examine XY models with plural spin sites (sites number L) inside a unit cell, and with two kinds of coupling constants, J and $-K$. One of the coupling constants, $-K$, belongs to the interaction between the nearest spins inside the same unit cell, and the other coupling constant J belongs to the interaction between the spin of the right end of a unit cell and the spin of the left end of the next unit cell.

In Sec. III, we will prove that this system has the following properties: there is an energy gap between the ground state and the first excited state for the system with an arbitrary even number of spin sites inside a unit cell, and there is no gap for the system with an arbitrary odd number of spin sites. Thus, in this paper we show the interesting relation between the spin-site number L and the existence of the energy gap. This relation in the XY models with plural spin sites is the same as that discussed in Haldane's conjecture, where it should be noted that the spins inside a unit cell construct the integral (or half integral) angular momentum for any even (or odd) number of spin sites inside a unit cell.

Here we examine investigations of spin chain models which were carried out hitherto [5–7]. Kontorovich and Tsukernik, Smith, Beni, and Pincus, Perk, Okamoto, and many other authors have examined XY models with two or three spin sites ($L=2$ and 3). Matsubara and Katsura, D. Cabib and Mahanti, and Braeter and Kowalski studied the XY model with random coupling constants [6]. These works showed that random interactions influence thermodynamic properties. Bonner and Blote, and many other authors, examined the spin-$\frac{1}{2}$ alternating Heisenberg chain, and calculated the excitation energy of the finite size chain [7].

These works did not prove the relation between the number of spin sites inside a unit cell and the gap in excitation energy spectra. Therefore, we point out this relation in this paper, and prove it in Sec. III.

In Sec. IV, we exactly calculate the magnetization and susceptibility of the present system. From the results, it becomes clear that the function form of the magnetization versus applied magnetic field H are classified into the following two types near $H=0$.

(1) First type: The magnetization smoothly increases with the increment of the magnetic field H. The susceptibility is positive at temperature $T=0$. This type appears in a system with any odd number of spin sites inside a unit cell.

(2) Second type: The magnetization is zero below the critical magnetic field H_{c1} at $T=0$, and steeply increases at $H>H_{c1}$. The susceptibility is zero at $T=0$. This second type belongs to a system with any even numbers of spin sites inside a unit cell.

The graphs of the magnetization versus H ($H>0$) has Q plateaus near zero temperature, where Q is the integer part of $(L+2)/2$. This complex dependence comes from the existence of band gaps in the eigenenergy levels. It may be interesting to compare the calculated results with several substances. There are many experiments [8,9] for $S=1$ linear Heisenberg antiferromagnets, for example $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ (NETP), tetramethyamine manganese trichloride (TMMC), and $(CH_3)_4NNi(NO_2)_3$ (TMNIN). NETP is the typical substance of the $S=1$

linear Heisenberg antiferromagnet, but the magnetization is measured only for a part of the region from zero to its magnetic saturation field. On the other hand, the magnetization of TMNIN is measured for the whole region of the magnetic field [9]. When applying a magnetic field H to TMNIN, the magnetization is very small for $H<H_{C1}$, but becomes large for $H>H_{C1}$ in a steep slope. The ratio of this critical magnetic field H_{C1} and the magnetic saturation field H_{C2} is very small (about $H_{C1}/H_{C2}\approx 1/10$). This behavior is different form the pure $S=1$ linear Heisenberg antiferromagnet. Therefore, it may be valuable to compare the data of TMNIN with our calculation result. Then, we obtain a good agreement except for the region of the magnetic field strength from 23 to 33 T (see Sec. IV). This fact shows that the substance TMNIN is different from a pure $S=1$ Heisenberg spin chain.

II. INTERACTION HAMILTONIAN

In this paper, we will examine the following Hamiltonian with two coupling constants J and $-K$. A Hamiltonian with L spin sites inside a unit cell is shown by

$$H_L=J\sum_{n=1,\ldots,N}(\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}+\sigma^{(y)}_{L,n}\sigma^{(y)}_{1,n+1}) -K\sum_{n=1,\ldots,N}\sum_{s=1,\ldots,L-1}(\sigma^{(x)}_{s,n}\sigma^{(x)}_{s+1,n}+\sigma^{(y)}_{s,n}\sigma^{(y)}_{s+1,n})-\sum_{n=1,\cdots,N}\sum_{s=1,\cdots,L}\tfrac{1}{2}g\mu_B H\sigma^{(z)}_{s,n}\ , \tag{2.1}$$

where H is the magnitude of the applied magnetic field in the z direction, g is the g value, and μ_B is the Bohr magneton. Also, the spin matrices have the well known form

$$\sigma^{(x)}=\begin{bmatrix}0&1\\1&0\end{bmatrix},\quad \sigma^{(y)}=\begin{bmatrix}0&-i\\i&0\end{bmatrix},\quad \sigma^{(z)}=\begin{bmatrix}1&0\\0&-1\end{bmatrix}. \tag{2.2}$$

The subscripts of the spin matrices $\sigma^{(x)}_{s,n}$, $\sigma^{(y)}_{s,n}$, and $\sigma^{(z)}_{s,n}$ denote the site number s and cell number n.

In order to diagonalize the Hamiltonian H_L, we transform the Hamiltonian H_L to a Hamiltonian H'_L. The transformation is the extension of the Jordan and Wigner transformation [10] for the case with plural spin sites (which is shown in Appendix A). From the results, the new Hamiltonian H'_L is given by

$$H'_L=2J\sum_p[e^{-ip}a^*_L(p)a_1(p)+e^{ip}a^*_1(p)a_L(p)] -2K\sum_p\sum_{s=1,\ldots,L-1}[a^*_s(p)a_{s+1}(p)+a^*_{s+1}(p)a_s(p)] -\sum_p\sum_{s=1,\ldots,L}g\mu_B Ha^*_s(p)a_s(p)+(L/2)g\mu_B HN\ , \tag{2.3}$$

where N is the maximum number of unit cells, and p is the wave number which takes such values as

$$p=(2\pi/N)\times(\text{integer}),\quad -\pi<p\le\pi\ . \tag{2.4}$$

In Eq. (2.3), the operators $a^*_s(p)$ and $a_s(p)$ are the creation and annihilation operators of the fermion with site numbers s and wave number p. These operators satisfy the well-known anticommutation relations, as follows:

$$\{a_s(p),a^*_t(q)\}=\delta_{s,t}\delta_{p,q}\ , \tag{2.5a}$$

$$\{a_s(p),a_t(q)\}=0,\quad \{a^*_s(p),a^*_t(q)\}=0\ . \tag{2.5b}$$

This Hamiltonian H'_L is the sum of the following operators $W(p)$:

$$W(p)=2J[e^{-ip}a^*_L(p)a_1(p)+e^{ip}a^*_1(p)a_L(p)] -2K\sum_{s=1,\ldots,L-1}[a^*_s(p)a_{s+1}(p)+a^*_{s+1}(p)a_s(p)] -g\mu_B H\sum_{s=1,\cdots,L}a^*_s(p)a_s(p)\ . \tag{2.6}$$

Since $[W(p),W(q)]=0$, we may solve the following eigenequation in order to diagonalize H'_L:

$$W_p\sum_{s=1,\ldots,L}\alpha_s(p)a^*_s(p)|0\rangle =[\lambda(p)-g\mu_B H]\times\sum_{s=1,\ldots,L}\alpha_s(p)a^*_s(p)|0\rangle\ , \tag{2.7}$$

where $\lambda(p)$ denotes the eigenenergy at the applied magnetic field $H=0$. Let us write this eigenequation into the matrix form as follows:

$$\mathbf{M}\boldsymbol{\alpha}=\lambda(p)\boldsymbol{\alpha}\ , \tag{2.8}$$

where the matrix $\mathbf{M}$ has the matrix elements

$$M_{1j}=-2K\delta_{2,j}\ \text{ for } 1\le j\le L-1,\quad M_{1L}=2Je^{ip}\ , \tag{2.9a}$$

$$M_{ij}=-2K(\delta_{i+1,j}+\delta_{i-1,j}),\ \text{ for } 2\le i\le L-1\ , \tag{2.9b}$$

$$M_{L1}=2Je^{-ip},\quad M_{Lj}=-2K\delta_{L-1,j}\ \text{ for } 2\le j\le L\ . \tag{2.9c}$$

This eigenequation has L solutions. The sth eigenvector and its eigenvalue are denoted by $\alpha^{(s)}(p)$ and $\lambda^{(s)}(p)$, respectively. By using these eigenvectors and eigenvalues, we obtain the diagonal form of the Hamiltonian H_L' as

$$H_L'=\sum_p \sum_{s=1,\ldots,L} [\lambda^{(s)}(p)-g\mu_B H][A_s(p)]^* A_s(p) + (L/2)g\mu_B HN , \tag{2.10}$$

where

$$[A_s(p)]^*=\sum_{t=1,\ldots,L} \alpha_t^{(s)}(p) a_t^*(p) , \tag{2.11}$$

and $A_s(p)$ is the hermite conjugate operator of $[A_s(p)]^*$. The operators $[A_s(p)]^*$ and $A_s(p)$ satisfy the anticommutation relations;

$$\{[A_s(p)]^*,[A_t(q)]^*\}=0, \quad \{A_s(p),A_t(q)\}=0 , \qquad \{A_s(p),[A_t(q)]^*\}=\delta_{s,t}\delta_{p,q} . \tag{2.12}$$

Thus Hamiltonian H_L' has been completely diagonalized as in Eq. (2.10). At the applied magnetic field $H=0$, the ground-state energy E_G becomes

$$E_G=\sum_s \left[\sum_{p \text{ for } \lambda^{(s)}(p)<0} \lambda^{(s)}(p) \right] , \tag{2.13}$$

and the ground state is

$$|\text{ground state}\rangle=\prod_s \left[\prod_{p \text{ for } \lambda^{(s)}(p)<0} A_s(p)^* \right] |0\rangle . \tag{2.14}$$

The excited state is produced by the annihilation of the fermion $A_s(p)$ for $\lambda^{(s)}(p)<0$ or by the creation of fermion $[A_s(p)]^*$ for $\lambda^{(s)}(p)>0$. Accordingly, the single excitation energy becomes $|\lambda^{(s)}(p)|$. Next we examine the eigenvalues of the Hamiltonian H_L' for $L=2$, 3, and 4.

Now we show the results of the diagonalization of H_2'. The case $L=2$ was already solved in Ref. [5]. The two eigenvalues of Eq. (2.8) for $L=2$ are easily obtained as

$$\lambda^{(1)}(p)=2\sqrt{J^2+K^2-2JK\cos(p)} , \qquad \lambda^{(2)}(p)=-2\sqrt{J^2+K^2-2JK\cos(p)} . \tag{2.15}$$

At $H=0$, the excited state is produced by the annihilation of the fermion $A_2(p)$ or by the creation of the fermion $[A_1(p)]^*$. Then the single excitation energy becomes $|\lambda^{(s)}(p)|$ ($s=1$ and 2). Since $\lambda^{(s)}(p)\neq 0$ for $|K/J|\neq 1$, an energy gap appears for any case other than $J=K$ or $J=-K$. We show the function forms of $\lambda^{(1)}(p)$ and $\lambda^{(2)}(p)$ in Fig. 1. Thus there is an energy gap for $|J|\neq|K|$.

Now we show results of the diagonalization of H_3'. The eigenvalue of H_3' is denoted by $\lambda(p)-g\mu_B H$. Then $\lambda(p)$ satisfies

$$-[\lambda(p)]^3+(4J^2+8K^2)\lambda(p)+16JK^2\cos p=0 . \tag{2.16}$$

The three solutions of Eq. (2.16) are denoted by $\lambda^{(s)}(p)$ ($s=1$, 2, and 3). We show the graphs of $\lambda^{(s)}(p)$ in Fig. 2.

At $p=\pm\pi/2$, $\lambda(p)=0$ is one of the solutions of Eq. (2.16) for any value of the coupling constants J and K.

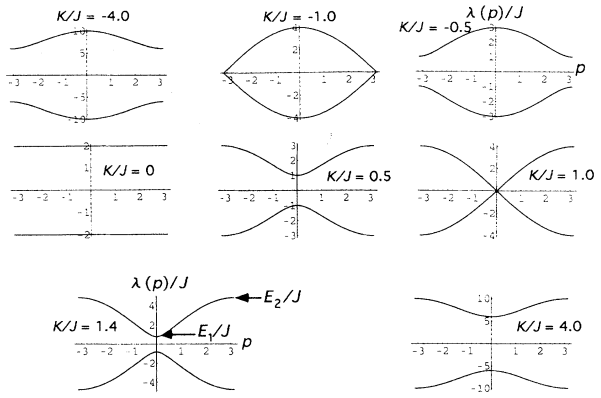

FIG. 1. Eigenenergies $\lambda^{(1)}(p)$ and $\lambda^{(2)}(p)$.

Therefore, there is no energy gap between the ground state and the first excited state for $H=0$.

Now we show results of the diagonalization of H_4'. When the eigenvalue of H_4' is denoted by $\lambda(p)-g\mu_B H$, $\lambda(p)$ satisfies

$$\begin{vmatrix} -\lambda(p) & -2K & 0 & 2Je^{ip} \\ -2K & -\lambda(p) & -2K & 0 \\ 0 & -2K & -\lambda(p) & -2K \\ 2Je^{-ip} & 0 & -2K & -\lambda(p) \end{vmatrix} =0 ,$$

which gives

$$\lambda(p)^4-(12K^2+4J^2)\lambda(p)^2+16K^4+32\cos(p)K^3J+16K^2J^2=0 . \tag{2.17}$$

The four solutions of Eq. (2.17) are denoted by $\lambda^{(s)}(p)$ ($s=1$, 2, 3, and 4). Then the solutions are

$$\lambda^{(1)}(p)=\sqrt{a(K,J)+\sqrt{a(K,J)^2-b(K,J)}} , \qquad \lambda^{(2)}(p)=-\sqrt{a(K,J)+\sqrt{a(K,J)^2-b(K,J)}} , \qquad \lambda^{(3)}(p)=\sqrt{a(K,J)-\sqrt{a(K,J)^2-b(K,J)}} , \qquad \lambda^{(4)}(p)=-\sqrt{a(K,J)-\sqrt{a(K,J)^2-b(K,J)}} , \tag{2.18}$$

where

$$a(K,J)=6K^2+2J^2 , \qquad b(K,J)=16[K^4+2\cos(p)K^3J+K^2J^2] . \tag{2.19}$$

We show the graphs of $\lambda^{(s)}(p)$ ($s=1$, 2, 3, and 4) in Fig. 3.

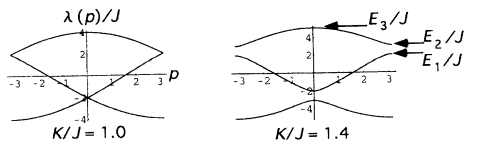

FIG. 2. Eigenenergies $\lambda^{(s)}(p)$ ($s=1$, 2, and 3).

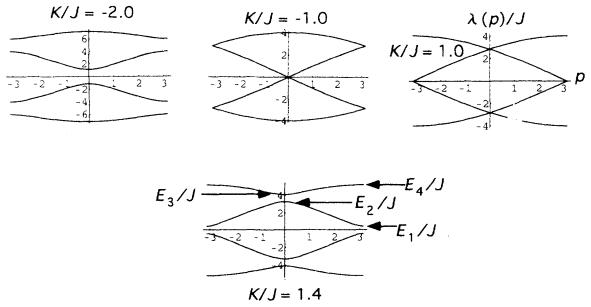

FIG. 3. Eigenenergies $\lambda^{(s)}(p)$ ($s=1$, 2, 3, and 4).

At $J=\pm K$, the gap disappears as in Fig. 3. Other cases, namely cases of $|J|\neq|K|$, have an energy gap between the ground state and the first excited state.

III. PROOF OF THE EXISTENCE OF AN ENERGY GAP FOR AN ARBITRARY EVEN NUMBER OF SPIN SITES

In the absence of the external magnetic field H, we will prove the existence of an energy gap for the system with any even number of spin sites inside a unit cell, and that there is no gap for the system with any odd number of spin sites inside a unit cell. As seen in Eq. (2.8), the eigenenergies of the present system for $H=0$ are the eigenvalues of the matrix $\mathbf{M}$, which has the matrix element M_{ij} as defined by Eq. (2.9). If the determinant of the matrix $\mathbf{M}$ becomes zero, the matrix has the eigenvalue of zero. Therefore, the excitation energy between the ground state and the first excited state becomes zero. That is to say there is no energy gap. On the other hand, if the determinant of the matrix $\mathbf{M}$ is not zero, the matrix does not have the eigenvalue zero. Therefore, there is an energy gap between the ground state and the first excited state.

Now we will calculate the determinant of the matrix $\mathbf{M}$,

$$\det\mathbf{M}=\begin{vmatrix} 0 & -2K & 0 & \cdots & 0 & 2Je^{ip} \\ -2K & 0 & -2K & \cdots & 0 & 0 \\ 0 & -2K & 0 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & 0 & -2K \\ 2Je^{-ip} & 0 & 0 & \cdots & -2K & 0 \end{vmatrix} ,$$

for the case of any even number of L. From the results of Appendix B, we can obtain the determinant for arbitrary even number of L as follows:

$$\det\mathbf{M}=(-1)^{L/2}(2K)^{L-2}[(|2K|-|2J|)^2+8|KJ| +(-1)^{-L/2}8KJ\cos p] . \quad (3.1)$$

In the case of $|K|\neq|J|$, the first term in the square bracket of Eq. (3.1) is positive, namely, $(|2K|-|2J|)^2>0$. The sum of the residual terms is also non-negative as

$$8|KJ|+(-1)^{-L/2}8KJ\cos p\geq 0 .$$

Therefore, $(\det\mathbf{M})$ is not zero for any wave number p. This means that any eigenvalue $\lambda(p)$ is not zero. That is to say, the minimum value of $|\lambda(p)|$ is positive. Since the matrix $\mathbf{M}$ has L rows and L columns, there are L eigenvalues, which are denoted by $\lambda^{(1)}(p)$, $\lambda^{(2)}(p)$,..., $\lambda^{(L)}(p)$. Using these eigenvalues, we can write the diagonal form of the Hamiltonian H'_L as

$$H'_L=\sum_{s=1}^{L}\sum_{p}[\lambda^{(s)}(p)A_s(p)^*A_s(p)] , \quad (3.2)$$

where we have substituted $H=0$ into Eq. (2.10). The ground state is such that the fermions occupy all the energy levels with negative eigenvalues. Therefore, the excitation energy between the ground state and the first excited state is the minimum value of $|\lambda(p)|$. This value is positive for the case of any even number of L and $|K|\neq|J|$. Consequently, the system has an energy gap between the ground state and the first excited state. When $|K|=|J|$, the energy gap disappears.

From Appendix B, for the case of any odd number of L, we obtain

$$\det\mathbf{M}=4J(2K)^{L-1}\cos p . \quad (3.3)$$

This result gives the fact that one eigenvalue becomes zero at $p=\pm\pi/2$. Therefore, there is no gap energy between the ground state and the first excited state for any odd number of spin sites inside a unit cell.

IV. MAGNETIZATION

In this section, we consider the canonical ensemble of the present system. We denote the number operator $[A_s(p)]^*A_s(p)$ by $n_s(p)$, and its mean value by $\bar{n}_s(p)$. We calculate the probabilities when $n_s(p)$ takes the eigenvalue 0 or 1. The probabilities are proportional to the following Boltzmann factors:

$$\exp(-0/k_BT)=1 \quad \text{for } n_s(p)=0 , \quad (4.1a)$$

$$\exp(-[\lambda^{(s)}(p)-g\mu_BH]/(k_BT)) \quad \text{for } n_s(p)=1 , \quad (4.1b)$$

where k_B is the Boltzmann's constant, and T is the temperature. Then these probabilities give the mean value $\bar{n}_s(p)$, as follows:

$$\bar{n}_s(p)=\exp[-\{\lambda^{(s)}(p)-g\mu_BH\}/(k_BT)]/\{1+\exp[-\{\lambda^{(s)}(p)-g\mu_BH\}/(k_BT)]\} . \quad (4.2)$$

Magnetization of the system $M(H,T)$ is described in terms of $\bar{n}_s(p)$ as follows:

$$M(H,T)=\tfrac{1}{2}g\mu_B\sum_{p}\sum_{s=1}^{L}[2\bar{n}_s(p)-1)] , \quad (4.3)$$

$$M(H,T)=\tfrac{1}{2}g\mu_B \times\sum_p\sum_{s=1}^{L}\{\exp[-\{\lambda^{(s)}(p)-g\mu_BH\}/(k_BT)]-1\}/\{1+\exp[-\{\lambda^{(s)}(p)-g\mu_BH\}/(k_BT)]\}$$

$$=\tfrac{1}{2}g\mu_B\sum_p\sum_{s=1}^{L}(-1)\tanh[\{\lambda^{(s)}(p)-g\mu_BH\}/(2k_BT)]\ , \tag{4.4}$$

$$M(H,T)=\tfrac{1}{2}g\mu_B(N/(2\pi))\int_{-\pi}^{\pi}\sum_{s=1}^{l}(-1)\tanh[\{\lambda^{(s)}(p)-g\mu_BH\}/(2k_BT)]dp\ . \tag{4.5}$$

The derivative of the magnetization by H is given as follows:

$$dM(H,T)/dH=\tfrac{1}{2}g\mu_BNg\mu_B/(4\pi k_BT)\int_{-\pi}^{\pi}\sum_{s=1}^{l}\mathrm{sech}^2[\{\lambda^{(s)}(p)-g\mu_BH\}/(2k_BT)]dp\ . \tag{4.6}$$

Now, we numerically calculate the magnetization $M(H,T)$ and its derivative $dM(H,T)/dH$ for the cases of $L=2$, 3, and 4. The calculation result for $L=2$ is shown in Fig. 4.

As seen in this figure, the calculated value of the magnetization has two plateaus of $0\le H\le H_{C1}$, and $H_{C2}\le H$. Moreover, $dM(H,T)/dH$ has two peaks near $H=H_{C1}$ and $H=H_{C2}$. At the temperature $T=0$, the magnetization is zero in the range of $0\le H\le H_{C1}$, and is constant in the range of $H_{C2}\le H$. Here, the critical values H_{C1} and H_{C2} are equal to $E_1/g\mu_B$ and $E_2/g\mu_B$, respectively, where E_1 and E_2 are shown in Fig. 1.

Now we want to compare experimental data with this calculation result. In the comparison, we need the data of the magnetization from the magnetic field zero to the saturated magnetic field. However, the author did not find a suitable substance with the type of interaction of the XY model.

Although the substance $(CH_3)_4NNi(NO_2)_3$ (TMNIN) has the same type of interaction as the XXZ model, the magnetization data of TMNIN are measured from the magnetic field zero to the saturated magnetic field [9].

Therefore, it may be valuable to compare the data with our calculation results (see Fig. 5). As can be easily seen from the figure, the calculated value of the magnetization agrees with the data of the magnetization of TMNIN except for 23–33 T. Thus the data show that a quenching of the gap appears around 2.7 T, and the value is very small in comparison with the magnetic saturation field of 33 T. This means that TMNIN is close to the $S=1$ XY model, but far from $S=1$ Heisenberg chain system.

Next, we calculate the magnetization and $dM(H,T)/dH$ for $L=3$ and 4. The results are shown in Figs. 6 and 7, respectively. Figure 6 shows that there are two plateaus of the magnetization in the ranges $H_{C1}\le H\le H_{C2}$ and $H_{C3}\le H$. Also, Fig. 7 shows that there are three plateaus of the magnetization in the ranges $0\le H\le H_{C1}$, $H_{C2}\le H\le H_{C3}$, and $H_{C4}\le H$. These plateaus correspond to the energy gaps. That is to say,

$$H_{Cs}=E_s/g\mu_B\quad(s=1,2,3\text{ or }s=1,2,3,4) \tag{4.7}$$

where E_s is shown in Figs. 2 and 3.

Finally, we calculate the temperature dependence of the susceptibility, namely $dM(H,T)/dH$ at $H=0$. The calculation results are shown in Fig. 8 for $L=2$, 3, and 4. As seen in this figure, the susceptibilities are zero at the temperature zero for $L=2$ and 4. On the other hand, for $L=3$, its susceptibility takes a positive value even at $T=0$. Thus, the susceptibility is zero at $T=0$ for any even number of the spin sites inside a unit cell because of an excitation energy gap between the ground state and the first excited state, but is positive at $T=0$ for any odd number of the spin sites inside a unit cell because there is

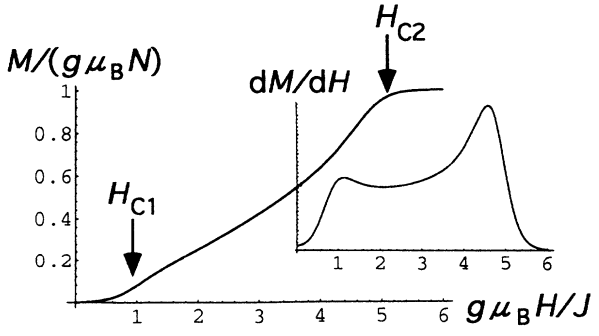

FIG. 4. Magnetization and $dM(H,T)/dH$ at $K/J=1.4$, and $k_BT/J=0.2$ for the system with two spin sites in a unit cell.

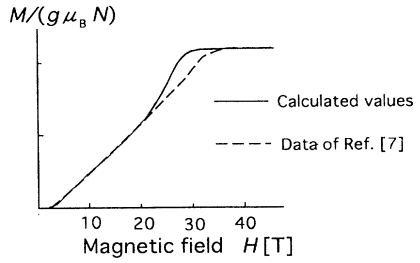

FIG. 5. The data of the magnetization of TMNIN, and the calculated values of the magnetization for $L=2$, $K/J=1.4$, and $k_BT/J=0.2$.

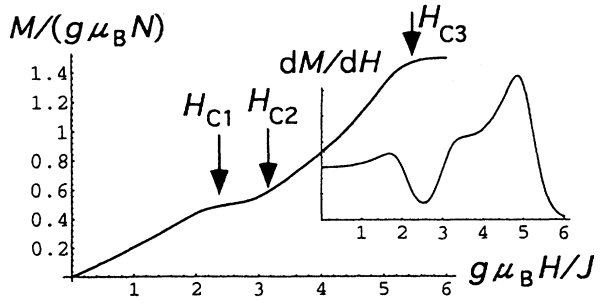

FIG. 6. Magnetization and $dM(H,T)/dH$ at $K/J=1.4$ and $k_BT/J=0.2$ for the system with three spin sites in a unit cell.

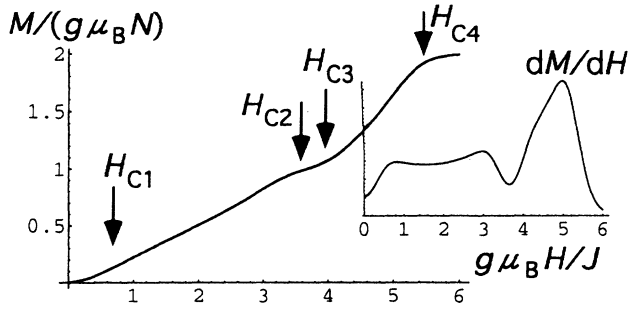

FIG. 7. Magnetization and $dM(H,T)/dH$ at $K/J=1.4$ and $k_BT/J=0.2$ for the system with four spin sites in a unit cell.

no gap in the excitation energy between the ground state and the first excited state.

V. CONCLUSIONS

In this paper we proved that the energy gap between the ground state and the first excited state appears for any even number of spin sites inside a unit cell, and that the energy gap disappears for any odd number of spin sites. At $T=0$, the magnetization is zero in the range of $0\leq H\leq H_{C1}$ for any even number of spin sites inside a unit cell, where $H_{C1}=E_1/g\mu_B$, and E_1 is the energy gap between the ground state and the first excited state for $H=0$. Therefore, the susceptibility is zero for any even number of L. On the other hand, for any odd number of L, the magnetization linearly increases from zero at $T=0$, and therefore the susceptibility is positive for any odd number of L. It has been found to be a property of the present system that the function form of the magnetization versus $H(H\geq 0)$ has Q plateaus at $T=0$, where Q is the integer part of $(L+2)/2$.

ACKNOWLEDGMENTS

I would like to express my heartfelt thanks to Professor A. Ikeda, Professor K. Katsumata, and Professor H. Hori. If they did not encourage me, this work would not have been completed. I would also like to express my heartfelt thanks to Professor K. Deguchi for use of his computer.

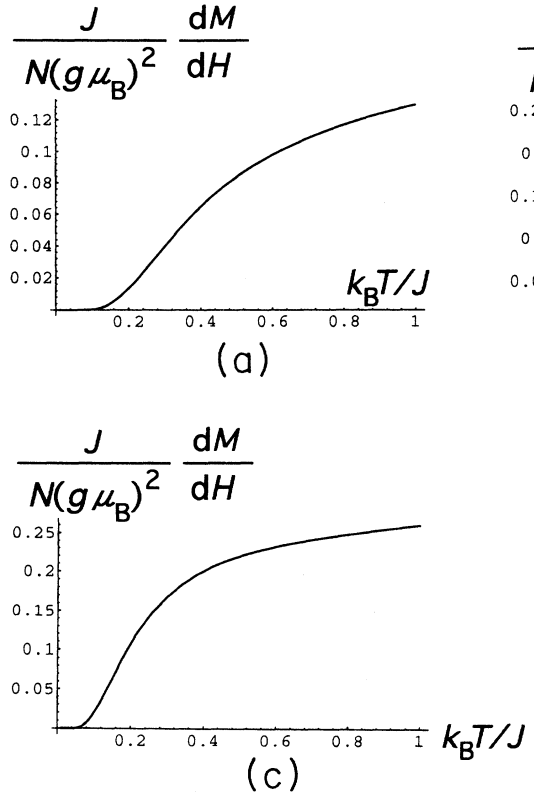

FIG. 8. (a) Susceptibility for $L=2$. (b) Susceptibility for $L=3$. (c) Susceptibility for $L=4$.

APPENDIX A

In order to diagonalize the Hamiltonian, we carry out the following isomorphic mapping from the spin states to fermion states. This mapping is an extension of the Jordan and Wigner transformation [10] into a case with plural spin sites. By mapping, an up-spin state is transformed to a one-fermion state, and a down-spin state is transferred to a zero-fermion state. Here we denote the fermion creation operator with site number s and cell number n by $a^*_{s,n}$. The fermion operators $a^*_{s,n}$ satisfy the well-known anticommutation relations as

$$\{a_{s,n},a^*_{t,m}\}=\delta_{s,t}\delta_{n,m},\{a_{s,n},a_{t,m}\}=0,\quad \{a^*_{s,n},a^*_{t,m}\}=0\ , \tag{A1}$$

where $\{a_{s,n},a^*_{t,m}\}=a_{s,n}a^*_{t,m}+a^*_{t,m}a_{s,n}$, and $\delta_{n,m}$ is the Kronecker delta function. In the mapped states, all the creation operators are arranged in the following order; we put $a^*_{s,n}$ to the left of $a^*_{t,m}$ for $n<m$, and put $a^*_{s,n}$ to the left of $a^*_{t,n}$ for the same cell numbers n and $s<t$.

An example for the case of $L=4$ is

$$\begin{array}{ll}\text{spin state} & \text{fermion state}\\ \uparrow\downarrow\uparrow\uparrow\cdots\downarrow\downarrow\downarrow\uparrow\rightarrow & a^*_{1,1}a^*_{3,1}a_{4,1}\cdots{}^*\cdot a^*_{4,N}|0\rangle\ ,\end{array}$$

where $|0\rangle$ is the vacuum state. Thus we have completed the mapping from the spin states to the fermion states. Next, the spin angular momentum operators in H_L are transformed as follows:

$$\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}\rightarrow X_{L,n;1,n+1}=(a^*_{L,n}-a_{L,n})(a^*_{1,n+1}+a_{1,n+1}), \tag{A2a}$$

$$\sigma^{(y)}_{L,n}\sigma^{(y)}_{1,n+1}\rightarrow Y_{L,n;1,n+1}=-(a^*_{L,n}+a_{L,n})(a^*_{1,n+1}-a_{1,n+1})\ , \tag{A2b}$$

$$\sigma^{(x)}_{s,n}\sigma^{(x)}_{s+1,n}\rightarrow X_{s,n;s+1,n}=(a^*_{s,n}-a_{s,n})(a^*_{s+1,n}+a_{s+1,n})\quad \text{for } 1\le s\le L-1\ , \tag{A2c}$$

$$\sigma^{(y)}_{s,n}\sigma^{(y)}_{s+1,n}\rightarrow Y_{s,n;s+1,n}=-(a^*_{s,n}+a_{s,n})(a^*_{s+1,n}-a_{s+1,n})\quad \text{for } 1\le s\le L-1\ , \tag{A2d}$$

$$\sigma^{(z)}_{s,n}\rightarrow Z_{s,n}=2a^*_{s,n}a_{s,n}-1\ . \tag{A2e}$$

Next, we show that these fermion operators $X_{L,n;1,n+1}$, $Y_{L,n;1,n+1}$, $X_{s,n;s+1,n}$, $Y_{s,n;s+1,n}$, and $Z_{s,n}$ have the same works as those of the operators $\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}$, $\sigma^{(y)}_{L,n}\sigma^{(y)}_{1,n+1}$, $\sigma^{(x)}_{s,n}\sigma^{(x)}_{s+1,n}$, $\sigma^{(y)}_{s,n}\sigma^{(y)}_{s+1,n}$, and $\sigma^{(z)}_{s,n}$, respectively. The proof is as follows. By multiplying the operator $\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}$ by any spin state, we obtain

$$\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}|\uparrow\uparrow\rangle=|\downarrow\downarrow\rangle\ , \tag{A3a}$$

$$\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}|\uparrow\downarrow\rangle=|\downarrow\uparrow\rangle\ , \tag{A3b}$$

$$\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}|\downarrow\uparrow\rangle=|\uparrow\downarrow\rangle\ , \tag{A3c}$$

$$\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}|\downarrow\downarrow\rangle=|\uparrow\uparrow\rangle\ , \tag{A3d}$$

where we have abbreviated all spin states other than the two spin states with site number L and cell number n, and site number 1 and cell number $n+1$. On the other hand, the operator $X_{L,n;1,n+1}$ has the properties

$$X_{L,n;1,n+1}a^*_{L,n}a^*_{1,n+1}|0\rangle=(a^*_{L,n}-a_{L,n})(a^*_{1,n+1}+a_{1,n+1})a^*_{L,n}a^*_{1,n+1}|0\rangle=|0\rangle\ , \tag{A4a}$$

$$X_{L,n;1,n+1}a^*_{L,n}|0\rangle=(a^*_{L,n}-a_{L,n})(a^*_{1,n+1}+a_{1,n+1})a^*_{L,n}|0\rangle=a^*_{1,n+1}|0\rangle\ , \tag{A4b}$$

$$X_{L,n;1,n+1}a^*_{1,n+1}|0\rangle=(a^*_{L,n}-a_{L,n})(a^*_{1,n+1}+a_{1,n+1})a^*_{1,n+1}|0\rangle=a^*_{L,n}|0\rangle\ , \tag{A4c}$$

$$X_{L,n;1,n+1}|0\rangle=(a^*_{L,n}-a_{L,n})(a^*_{1,n+1}+a_{1,n+1})|0\rangle=a^*_{L,n}a^*_{1,n+1}|0\rangle\ . \tag{A4d}$$

Therefore, the operator $X_{L,n;1,n+1}$ is equivalent to the operator $\sigma^{(x)}_{L,n}\sigma^{(x)}_{1,n+1}$. Next, we calculate the products of $Y_{L,n;1,n+1}$ and the fermion states

$$Y_{L,n;1,n+1}a^*_{L,n}a^*_{1,n+1}|0\rangle=-(a^*_{L,n}+a_{L,n})(a^*_{1,n+1}-a_{1,n+1})a^*_{L,n}a^*_{1,n+1}|0\rangle=-|0\rangle\ , \tag{A5a}$$

$$Y_{L,n;1,n+1}a^*_{L,n}|0\rangle=-(a^*_{L,n}+a_{L,n})(a^*_{1,n+1}-a_{1,n+1})a^*_{L,n}|0\rangle=a^*_{1,n+1}|0\rangle\ , \tag{A5b}$$

$$Y_{L,n;1,n+1}a^*_{1,n+1}|0\rangle=-(a^*_{L,n}+a_{L,n})(a^*_{1,n+1}-a_{1,n+1})a^*_{1,n+1}|0\rangle=a^*_{L,n}|0\rangle\ , \tag{A5c}$$

$$Y_{L,n;1,n+1}|0\rangle=-(a^*_{L,n}+a_{L,n})(a^*_{1,n+1}-a_{1,n+1})|0\rangle=-a^*_{L,n}a^*_{l,n+1}|0\rangle\ , \tag{A5d}$$

which are equivalent to the following relations:

$$\sigma^{(y)}_{L,n}\sigma^{(y)}_{1,n+1}|\uparrow\uparrow\rangle=-|\downarrow\downarrow\rangle\ , \tag{A6a}$$

$$\sigma^{(y)}_{L,n}\sigma^{(y)}_{1,n+1}|\uparrow\downarrow\rangle=|\downarrow\uparrow\rangle\ , \tag{A6b}$$

$$\sigma^{(y)}_{L,n}\sigma^{(y)}_{1,n+1}|\downarrow\uparrow\rangle=|\uparrow\downarrow\rangle\ , \tag{A6c}$$

$$\sigma^{(y)}_{L,n}\sigma^{(y)}_{1,n+1}|\downarrow\downarrow\rangle=-|\uparrow\uparrow\rangle\ . \tag{A6d}$$

Similarly, we can easily show the equivalence of the fer-

mion operators ($X_{s,n;s+1,n}$ and $Y_{s,n;s+1,n}$) and the operators ($\sigma^{(x)}_{s,n}\sigma^{(x)}_{s+1,n}$ and $\sigma^{(y)}_{s,n}\sigma^{(y)}_{s+1,n}$), respectively. Finally, we can verify that the fermion operator $Z_{s,n}$ does the same work as the spin operator $\sigma^{(z)}_{s,n}$, as follows:

$$Z_{s,n}a^*_{s,n}|0\rangle=(2a^*_{s,n}a_{s,n}-1)a^*_{s,n}|0\rangle=a^*_{s,n}|0\rangle\ , \tag{A7a}$$

$$Z_{s,n}|0\rangle=(2a^*_{s,n}a_{s,n}-1)|0\rangle=-|0\rangle\ , \tag{A7b}$$

$$\sigma^{(z)}_{s,n}|\uparrow\rangle=|\uparrow\rangle\ , \tag{A7c}$$

$$\sigma^{(z)}_{s,n}|\downarrow\rangle=-|\downarrow\rangle\ . \tag{A7d}$$

In the proofs mentioned above, we have treated only the nearest neighbor two spins. Therefore, it is necessary to show the commutability between these operators and the fermion operators of other sites, as follows:

$$[X_{L,n;1,n+1},a^*_{t,m}]=0 \text{ for } (t,m)\neq(L,n) \text{ and } (t,m)\neq(1,n+1)\ , \tag{A8a}$$

$$[X_{s,n;s+1,n},a_{t,m}]=0 \text{ for } (t,m)\neq(s,n) \text{ and } (t,m)\neq(s+1,n), \tag{A8b}$$

$$[Y_{L,n;1,n+1},a^*_{t,m}]=0 \text{ for } (t,m)\neq(L,n) \text{ and } (t,m)\neq(1,n+1)\ , \tag{A8c}$$

$$[Y_{s,n;s+1,n},a_{t,m}]=0 \text{ for } (t,m)\neq(s,n) \text{ and } (t,m)\neq(s+1,n)\ , \tag{A8d}$$

$$[Z_{s,n},a^*_{t,m}]=0 \text{ for } (t,m)\neq(s,n)\ . \tag{A8e}$$

Since these commutabilities are easily verified, we can conclude that the replacements (A2a)–(A2e) are the isomorphic mapping.

We transform the original Hamiltonian (2.1) by using the mapping (A2a)–(A2e) and then obtain the following Hamiltonian H'_L:

$$H'_L=2J\sum_{n=1,\ldots,N}(a^*_{L,n}a_{1,n+1}+a^*_{1,n+1}a_{L,n})-2K\sum_{n=1,\ldots,N}\sum_{s=1,\ldots,L-1}(a^*_{s,n}a_{s+1,n}+a^*_{s+1,n}a_{s,n})$$
$$-\sum_{n=1,..,N}\sum_{s=1,\ldots,L}\tfrac{1}{2}g\mu_B H(2a^*_{s,n}a_{s,n}-1)\ . \tag{A9}$$

The Hamiltonian H'_L has the same energy spectra as that of H_L because the mapping is isomorphic. In order to diagonalize the Hamiltonians H'_L, we introduce the Fourier transformation of the fermion operators,

$$a_{s,n}=(1/N)^{1/2}\sum_p e^{-ipn}a_s(p)\ , \tag{A10}$$

where N is the maximum number of unit cells and p is the wave number which takes such values as

$$p=(2\pi/N)\times(\text{integer}),\quad -\pi<p\leq\pi\ . \tag{A11}$$

We substitute Eq. (A10) and its hermite conjugate equation into the Hamiltonian (A9), and use the periodic boundary conditions

$$a_{1,N+1}=a_{1,1} \text{ and } a^*_{1,N+1}=a^*_{1,1} \tag{A12}$$

and also the property

$$(1/N)\sum_{n=1,..,N}e^{ipn}e^{-iqn}=\delta_{p,q}\ . \tag{A13}$$

Then we obtain

$$H'_L=2J\sum_p[e^{-ip}a^*_L(p)a_1(p)+e^{ip}a^*_1(p)a_L(p)]-2K\sum_p\sum_{s=1,\ldots,L-1}[a^*_s(p)a_{s+1}(p)+a^*_{s+1}(p)a_s(p)]$$
$$-\sum_p\sum_{s=1,\ldots,L}g\mu_B Ha^*_s(p)a_s(p)+\tfrac{1}{2}g\mu_B HNL\ . \tag{A14}$$

Thus Hamiltonian H_L is transformed into the bilinear form of the fermion operators in momentum space.

APPENDIX B

$$\det\mathbf{M}=\begin{vmatrix} 0 & -2K & 0 & \cdots & 0 & 2Je^{ip} \\ -2K & 0 & -2K & \cdots & 0 & 0 \\ 0 & -2K & 0 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \ddots \\ 0 & 0 & 0 & \cdots & 0 & -2K \\ 2Je^{-ip} & 0 & 0 & \cdots & -2K & 0 \end{vmatrix}, \tag{B1}$$

where the matrix $\mathbf{M}$ has L rows and L columns

$$\det\mathbf{M}=\sum_{j=1}^{L} M_{1j}A_{1j}, \tag{B2}$$

where A_{1j} denotes the cofactor of the element M_{1j}. Then we obtain

$$\det\mathbf{M}=-2KA_{12}+2Je^{ip}A_{1L}, \tag{B3}$$

$$A_{12}=-\begin{vmatrix} -2K & -2K & 0 & \cdots & 0 & 0 \\ 0 & 0 & -2K & \cdots & 0 & 0 \\ 0 & -2K & 0 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & 0 & -2K \\ 2Je^{-ip} & 0 & 0 & \cdots & -2K & 0 \end{vmatrix}, \tag{B4}$$

$$A_{1L}=(-1)^{L+1}\begin{vmatrix} -2K & 0 & -2K & \cdots & 0 & 0 \\ 0 & -2K & 0 & \cdots & 0 & 0 \\ 0 & 0 & -2K & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & -2K & 0 \\ 2Je^{-ip} & 0 & 0 & \cdots & 0 & -2K \end{vmatrix}, \tag{B5}$$

where it should be noted that the matrices appearing on the right-hand sides of Eqs. (B4) and (B5) have $L-1$ rows and $L-1$ columns. We denote the cofactor of the (i,j) element of the matrix appearing in Eq. (B4) by B_{ij}; then the cofactor A_{12} becomes

$$A_{12}=2KB_{11}-2Je^{-ip}B_{L-1,1}, \tag{B6}$$

where

$$B_{11}=\begin{vmatrix} 0 & -2K & 0 & \cdots & 0 & 0 \\ -2K & 0 & -2K & \cdots & 0 & 0 \\ 0 & -2K & 0 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & 0 & -2K \\ 0 & 0 & 0 & \cdots & -2K & 0 \end{vmatrix}$$

$$=\begin{cases} (-1)^{(L-2)/2}(2K)^{L-2} & \text{for an even number of } L \\ 0 & \text{for an odd number of } L \end{cases}, \tag{B7}$$

$$B_{L-1,1}=(-1)^{L}\begin{vmatrix} -2K & 0 & 0 & \cdots & 0 & 0 \\ 0 & -2K & 0 & \cdots & 0 & 0 \\ -2K & 0 & -2K & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & -2K & 0 \\ 0 & 0 & 0 & \cdots & 0 & -2K \end{vmatrix}$$

$$=(-1)^{L}(-2K)^{L-2}=(2K)^{L-2}, \tag{B8}$$

where it should be noted here that the matrices appearing in the middle sides of Eqs. (B7) and (B8) have $L-2$ rows and $L-2$ columns. Substitution of Eqs. (B7) and (B8) into (B6) gives

$$A_{12}=\begin{cases}(-1)^{(L-2)/2}(2K)^{L-1}-2Je^{-ip}(2K)^{L-2} & \text{for an even number of } L\\ -2Je^{-ip}(2K)^{L-2} & \text{for an odd number of } L\ .\end{cases} \tag{B9}$$

Similarly, we denote the cofactor of the (i,j) element of the matrix appearing in Eq. (B5) by C_{ij}; then the cofactor A_{1L} becomes

$$A_{1L}=(-1)^{L+1}(-2KC_{11}+2Je^{-ip}C_{L-1,1})\ , \tag{B10}$$

$$C_{11}=\begin{vmatrix}-2K & 0 & -2K & \cdots & 0 & 0\\ 0 & -2K & 0 & \cdots & 0 & 0\\ 0 & 0 & -2K & \cdots & 0 & 0\\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots\\ 0 & 0 & 0 & \cdots & -2K & 0\\ 0 & 0 & 0 & \cdots & 0 & -2K\end{vmatrix}$$

$$=(-2K)^{L-2}\ , \tag{B11}$$

$$C_{L-1,1}=(-1)^{L}\begin{vmatrix}0 & -2K & 0 & \cdots & 0 & 0\\ -2K & 0 & -2K & \cdots & 0 & 0\\ 0 & -2K & 0 & \cdots & 0 & 0\\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots\\ 0 & 0 & 0 & \cdots & 0 & -2K\\ 0 & 0 & 0 & \cdots & -2K & 0\end{vmatrix}$$

$$=\begin{cases}(-1)^{(L-2)/2}(2K)^{L-2} & \text{for an even number of } L\\ 0 \text{ for an odd number of } L\end{cases} \tag{B12}$$

where it should be noted here that the matrices appearing in the middle sides of Eqs. (B11) and (B12) have $L-2$ rows and $L-2$ columns. Substitution of Eqs. (B11) and (B12) into (B10) gives

$$A_{1L}=\begin{cases}(-1)^{L+1}[(-2K)^{L-1}+2Je^{-ip}(-1)^{(L-2)/2}(2K)^{L-2}]\text{for an even number of } L\\ (2K)^{L-1} \text{ for an odd number of } L\ .\end{cases} \tag{B13}$$

By substituting Eqs. (B9) and (B13) into (B3), we obtain

$$\begin{aligned}\det\mathbf{M}&=-2K[(-1)^{(L-2)/2}(2K)^{L-1}-2Je^{-ip}(2K)^{L-2}]\\ &\quad+2Je^{ip}(-1)^{L+1}[(-2K)^{L-1}+2Je^{-ip}(-1)^{(L-2)/2}(2K)^{L-2}]\\ &=(-1)^{L/2}[(2K)^{L}+(2J)^{2}(2K)^{L-2}]+2J(2K)^{L-1}(e^{ip}+e^{-ip})]\\ &=(-1)^{L/2}(2K)^{L-2}[(|2K|-|2J|)^{2}+8|KJ|+(-1)^{-L/2}8KJ\cos p]\end{aligned}$$

(for an even number of L) . (B14)

Similarly, for an odd number of L, the determinant of the matrix $\mathbf{M}$ is

$$\begin{aligned}\det\mathbf{M}&=2K2Je^{-ip}(2K)^{L-2}+2Je^{ip}(2K)^{L-1}\\ &=2J(2K)^{L-1}(e^{ip}+e^{-ip})=4J(2K)^{L-1}\cos p \quad (\text{for an odd number of})L\ .\end{aligned} \tag{B15}$$

These results show the gap or gapless mechanism of the excitation energies.

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