The spin-Peierls phase transition in the quasi-one-dimensional spin-1/2 Heisenberg model

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Abstract. The spin-Peierls (SP) phase transition of the inorganic compound $CuGeO_3$ is theoretically studied on the basis of Heisenberg model using Jordan-Wigner transformation (JWT) by mean-field approach. The magnetic interaction is considered to be the quasi-one-dimensional Cu^{2+} (spin-1/2) Heisenberg chain, and the spin-lattice distortion is expressed quasiclassically. We found that the interchain coupling $J_\perp$ between the chains leads to the occurrence of gapless dimerization (or stepped SP) phase. As the interchain coupling decreases, the difference between the dimerization temperature t_D and the gap-opening temperature t_G decreases. If the interchain coupling is small enough, then the difference should be too small to be observed experimentally.

PACS. 75.10.Jm Quantized spin models – 75.30.Gw Magnetic anisotropy – 75.50.Ee Antiferromagnetics

1 Introduction

The discovery of the so-called spin-Peierls (SP) transition in a linear Cu^{2+} $(S = 1/2)$ chain inorganic compound $CuGeO_3$ [1] has attracted much attention both experimentally and theoretically [2–5]. The spin-Peierls transition can be interpreted as the formation of spin singlet due to the dimerization of localized d electrons of Cu^{2+}. The main feature of the state is the appearance of an energy gap between a nonmagnetic singlet ground state (GS) and a magnetic excited triplet state. Pouget et al. [6] observed an anomalous behavior of superstructure reflections indexed in a $2a \times b \times 2c$ lattice below the spin-Peierls transition temperature T_{sp} (14 K), and concluded that $CuGeO_3$ undergoes a second-order structural phase transition concomitantly to the disappearance of the magnetic susceptibility. Furthermore, a sharp change of specific heat [7,8] at T_{sp} confirms the appearance of a spin gap. The investigation of nonmagnetic impurity Zn-doping on the spectrum and dynamical spin structure factor of $CuGeO_3$ shows that the spin gap is rapidly suppressed by doping and the new phase is a three-dimensional (3D) Néel state [9,10].

Theoretical investigations of the SP transition have been performed on the basis of Heisenberg and quantum XY-models. A fermion representation via the Jordan-Wigner transformation (JWT) is used to describe the spin-1/2 chain, and the spin-phonon interactions are considered as the random-phase approximation [10–13].

Those theories only consider the intrachain magnetic coupling and neglect the interchain coupling which can lead the dimerization and the opening gap at the same temperature. However, Nishi et al. [14] reported the ratio of the interchain coupling $J_\perp$ to the intrachain coupling J is about 0.1. Therefore, the interchain coupling should be considered in theoretical calculations. In this paper, by considering the contribution of interchain coupling we propose a method to investigate the SP transition based on a quasi-one-dimensional Heisenberg model using JWT in the framework of the mean-field approach. We present two self-consistent equations for describing the variations of dimerization and interaction order parameters. It was found theoretically that the gapless spin-Peierls transition temperature is higher than that of the opening of the spin gap.

2 Model

One can consider a two-dimensional (2D) anisotropic Heisenberg model on a square lattice. The spin Hamiltonian is expressed as:

$$
\begin{aligned}
H &= J\sum_{i,j} S_{i,j}S_{i+\hat{x},j} + J_\perp \sum_{i,j} S_{i,j}S_{i,j+\hat{y}} \\
&= J\sum_{i,j}\{S_{i,j}^x S_{i+\hat{x},j}^x + S_{i,j}^y S_{i+\hat{x},j}^y + S_{i,j}^z S_{i+\hat{x},j}^z\} \\
&\quad + J_\perp \sum_{i,j}\{S_{i,j}^x S_{i,j+\hat{y}}^x + S_{i,j}^y S_{i,j+\hat{y}}^y + S_{i,j}^z S_{i,j+\hat{y}}^z\}
\end{aligned}
$$

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$$= J \sum_{i,j} \{\frac{1}{2}(S^+_{i,j}S^-_{i+\hat{x},j} + S^-_{i,j}S^+_{i+\hat{x},j}) + S^z_{i,j}S^z_{i+\hat{x},j}\}$$

$$+ J_\perp \sum_{i,j} \{\frac{1}{2}(S^+_{i,j}S^-_{i,j+\hat{y}} + S^-_{i,j}S^+_{i,j+\hat{y}}) + S^z_{i,j}S^z_{i,j+\hat{y}}\}, \quad (1)$$

where $S_{i,j}$ is the spin-1/2 operator at $S_{i,j} = i\hat{x} + j\hat{y}$, $\hat{x}$ and $\hat{y}$ are the unit vectors along the x-axis and y-axis, J is the intrachain exchange integral along the x-axis, and $J_\perp$ is the interchain exchange integral along the y-axis. There is no exact method for mapping the 2D spin-1/2 operators onto the fermion representation *via* the conventional JWT because the fermion operators defined on different chains are in general commuting instead of anticommuting. However, one can promote the conventional JWT to the 2D case by using some approximative schemes [15,16]. Here we use the transformation adopted by Azzouz [15], which is defined on a square lattice as

$$S^-_{i,j} = c_{i,j} \exp\{i\phi_{i,j}\}, \qquad (2)$$

$$S^+_{i,j} = \exp\{i\phi_{i,j}\}\, c^+_{i,j}, \qquad (3)$$

$$\phi_{i,j} = \pi \left[\sum_{d=0}^{i-1} \sum_{f=0}^{\infty} \hat{n}_{d,f} + \sum_{f=0}^{j-1} \hat{n}_{i,f} \right], \qquad (4)$$

where $c_{i,j}$ is a spinless fermion annihilation operator at $\hat{r}_{i,j} = i\hat{x} + j\hat{y}$, and $\hat{n}_{i,j} = c^+_{ij}c_{i,j}$ is the number operator at $\hat{r}_{i,j}$. Thus, the on-site exclusion principle of spin and the commutation relations are preserved. After substituting the extended JWT into the Hamiltonian (1), we have

$$H =$$
$$J \sum_{i,j} \{\frac{1}{2}(e^{i\varphi_{i,i+\hat{x},j}}c^+_{i,j}c_{i+\hat{x},j} + h.c.) + c^+_{i,j}c^+_{i+\hat{x},j}c_{i+\hat{x},j}c_{i,j}\}$$

$$+ J_\perp \sum_{i,j} \{\frac{1}{2}(c^+_{i,j}c_{i,j+\hat{y}} + h.c.) + c^+_{i,j}c^+_{i,j+\hat{y}}c_{i,j+\hat{y}}c_{i,j}\}, \quad (5)$$

where $e^{i\varphi_{i,j+\hat{x};j}} = e^{i[(\phi_{i+\hat{x},j}-\phi_{i,j})-\pi\hat{n}_{i,j}]}$ is a c-number. In equation (5), only the effective hopping between the nearest sites is taken into account for spinless fermion, in which J is in the x-direction (parallel to the chain), and $J_\perp$ is in the y-direction (perpendicular to the chain). The phase factor $e^{i\varphi_{i,i+\hat{x},j}}$ is determined by various spin configurations. Thus, similar to the in-phase flux state [17], each thick line corresponds to a phase factor of 1, and each thin line corresponds to a phase factor of $e^{\pm i\pi}$, as shown in Figure 1. This arrangement is such that each of the elementary plaquettes encloses a net flux of half-quanta. It is known that the spin dimerization is one of the most prominent features of the spin-Peierls state. According to the study of elastic neutron scattering for $CuGeO_3$ [18], we choose that in Figure 1 each thick and thin line along the x-axis corresponds to the stronger coupling $J_+ = J_0(1 + 2\delta u)$ and weaker coupling $J_- = J_0(1 - 2\delta u)$ respectively, while each thin line along the y-axis represents the weak coupling $J_\perp = \alpha J_0$. Here J_0 is the exchange integral along the x-axis before dimerization and $\delta = -dJ^x_{i,j}/du$. The
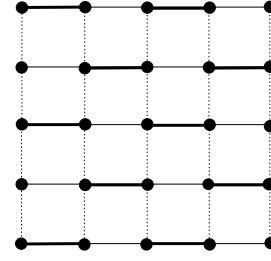

Fig. 1. Dimerization scheme on a square lattice, each thick and thin line along x-axis represents stronger coupling and weaker coupling respectively, the dotted line represents the interchain coupling; the phase distribution: each thick line corresponds to a phase factor of 1 and thin line corresponds to a phase factor of $e^{\pm i\pi} = -1$.

spin-lattice distortion u is treated quasiclassically as Su, Schrieffer and Heeger (SSH) [19] did for linear conducting chains. Following these descriptions, the dimerized Hamiltonian can be written in terms of spinless fermion operators a and b corresponding to the two sublattices A and B respectively, as follows:

$$H = \sum_{r_{i,j} \in A} \{-J_- \left[\frac{1}{2}(a^+_{i,j}b^+_{i-\hat{x}} + h.c.) - a^+_{i,j}b^+_{i-\hat{x},j}b_{i-\hat{x},j}a_{i,j} \right]$$

$$+ J_+ \left[\frac{1}{2}(a^+_{i,j}b_{i+\hat{x},j} + h.c.) + a^+_{i,j}b^+_{i+\hat{x},j}b_{i+\hat{x},j}a_{i,j} \right]$$

$$+ \alpha J_0 \left[\frac{1}{2}(a^+_{i,j}b_{i,j-\hat{y}} + a^+_{i,j}b_{i,j+\hat{y}} + h.c.) \right.$$

$$\left. + a^+_{i,j}b^+_{i,j+\hat{y}}b_{i,j+\hat{y}}a_{i,j} + a^+_{i,j}b^+_{i,j-\hat{y}}b_{i,j-\hat{y}}a_{i,j} \right]\}$$

$$+ \frac{1}{2} \sum_{r_{i,j} \in A} K[(u_{i,j} - u_{i-\hat{x},j})^2 + (u_{i+\hat{x},j} - u_{i,j})^2], \quad (6)$$

where $u_{i,j} = (-1)^{i+j}u$ is the spin-lattice distortion, and K is the elastic constant. The neutron scattering study [20] revealed that the distance of the dimerized Cu–Cu pairs is about 2.930 Å, and the interval of the two dimers is 2.955 Å. Although u is quite small as compared with the lattice spacing a, the quantity of $2J_0\delta u$ is not very small as compared with J_0, which should not be negligible. Therefore, the Hamiltonian (6) can be expressed in momentum space as

$$H = 2J_0 \sum_k (\gamma_k a^+_k b_k + h.c.)$$

$$+ \frac{2J_0}{N} \sum_{k,p,q} \xi_q a^+_k b^+_p b_{p+q}a_{k-q} + 2NKu^2, \qquad (7)$$

where

$$\gamma_k = \frac{1}{2}[2\delta u \cos(k_x a) + \alpha \cos(k_y a) + i \sin(k_x a)], \qquad (8)$$

$$\xi_k = \frac{1}{2}[\cos(k_x a) + \alpha \cos(k_y a) + i2\delta u \cos(k_x a)]. \qquad (9)$$

In equation (7), the four-fermion operator term can be decoupled in a Hartree-Fock approximation by introducing

$$E_k^\alpha = -\sqrt{J_0^2 c^2 \sin^2(k_x a) + J_0^2 [(2\delta u + P)\cos(k_x a) + \alpha(1+P)\cos(k_y a)]^2}, \tag{11}$$

$$E_k^\beta = \sqrt{J_0^2 c^2 \sin^2(k_x a) + J_0^2 [(2\delta u + P)\cos(k_x a) + \alpha(1+P)\cos(k_y a)]^2}, \tag{12}$$

an alternating order parameter $P = \langle a_0^+ b_0 \rangle / N$ determined numerically by self-consistent equations later. Thus, the Hamiltonian can be diagonalized as

$$H = \sum_k (E_k^\alpha \alpha_k^+ \alpha_k + E_k^\beta \beta_k^+ \beta_k) + N(1+\alpha)P^2 + 2NKu^2, \tag{10}$$

where

see equations (11, 12) above

here $c = (1 + 2\delta u P)$. It is convenient for the following calculations to set some parameters: $J_0 = 1$, $a = 1$, the reduced temperature $t = k_B T / J_0$, the dimerization parameter $\eta = \left| \frac{J_- - J_+}{J_+ + J_-} \right|$, and the reduced elastic constant $K' = K(J_0/\delta)^2$. Thus the reduced energy spectra can be written as:

$$\varepsilon_k^\alpha = -\sqrt{(1+\eta P)^2 \sin^2 k_x + [(\eta + P)\cos k_x + \alpha(1+P)\cos k_y]^2}, \tag{13}$$

$$\varepsilon_k^\beta = \sqrt{(1+\eta P)^2 \sin^2 k_x + [(\eta + P)\cos k_x + \alpha(1+P)\cos k_y]^2}. \tag{14}$$

From the spectra formula, the energy gap can be obtained as:

$$\Delta = 2[\eta + P - \alpha(1+P)]. \tag{15}$$

The reduced free energy per unit cell can be obtained from the diagonalized Hamiltonian (10), namely,

$$
\begin{aligned}
f &= -\frac{2}{J_0 N} k_B T \ln Z = -\frac{2t}{N} \ln(Tr e^{-H/k_B T}) \\
&= -\frac{2t}{N} \sum_{BZ'} [\ln(2\cosh \frac{E_k^\alpha}{2t}) + \ln(2\cosh \frac{E_k^\beta}{2t})] \\
&\quad + 2(1+\alpha)P^2 + K'\eta^2 \\
&= -t \iint_{BZ'} \frac{dk_x dk_y}{2\pi^2} [\ln(2\cosh \frac{E_k^\beta}{2t}) + \ln(2\cosh \frac{E_k^\beta}{2t})] \\
&\quad + 2(1+\alpha)P^2 + K'\eta^2.
\end{aligned}
\tag{16}
$$

Then, the dimerization and the alternating order parameters $\eta(t)$ and $P(t)$ can be determined in self-consistent way from the conditions: $\partial f / \partial \eta = 0$ and $\partial f / \partial P = 0$, as follows:

$$
\begin{aligned}
K' &= \iint_{BZ'} \frac{dk_x dk_y}{8\pi^2} \frac{PA\sin k_x + B\cos k_x}{\eta \sqrt{A^2 + B^2}} \\
&\quad \times [\tanh \frac{\varepsilon_k^\beta}{2t} - \tanh \frac{\varepsilon_k^\alpha}{2t}],
\end{aligned}
\tag{17}
$$

$$
\begin{aligned}
P &= \iint_{BZ'} \frac{dk_x dk_y}{8\pi^2} \frac{\eta A\sin k_x + B(\cos k_x + \alpha \cos k_y)}{2(1+\alpha)\sqrt{A^2 + B^2}} \\
&\quad \times [\tanh \frac{\varepsilon_k^\beta}{2t} - \tanh \frac{\varepsilon_k^\alpha}{2t}],
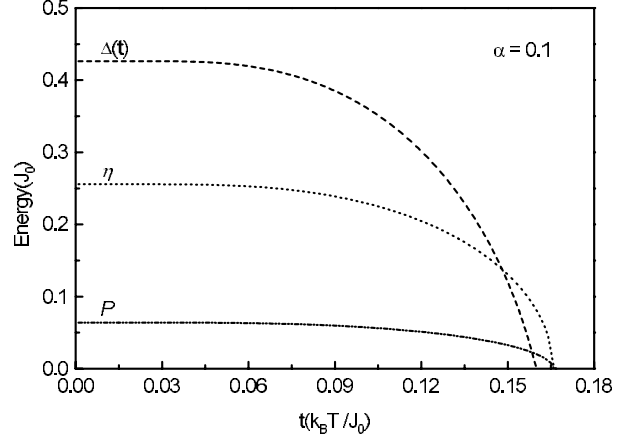\end{aligned}
\tag{18}
$$

Fig. 2. Temperature dependencies of the dimerization order parameter $\eta(t)$, the alternating order parameter $P(t)$ and the energy gap $\Delta(t)$ for $\alpha = 0.1$ and $K' = 0.708$.

where

$$A = (1 + \eta P)\sin k_x,$$
$$B = (\eta + P)\cos k_x + \alpha(1+P)\cos k_y.$$

The numerical results for the dimerization order parameter $\eta(t)$, the alternating order parameter $P(t)$ and the energy gap $\Delta(t)$ *versus* temperature are shown in Figure 2 by setting $\alpha = 0.1$. One can see that the dimerization and the opening of the gap happen at different temperatures, indicated by t_D (for $\eta = 0$) and t_G (for $\Delta = 0$), respectively. Figure 3 shows the variation of the energy spectrum from equation (14). It was found that for appropriately small order parameters ($\eta \leq 0.081$, $P \leq 0.0203$) the gapless state exists, and the dispersion relation along $O \to Y$ and $X \to O$ coincides with the experimental results for $CuGeO_3$ (see Ref. [17]). Such a dispersion reveals the existence of relatively strong *antiferromagnetic* coupling along the b axis [20].

From equations (17, 18), we can get the dimerization temperature t_D and the gap-opening temperature t_G numerically by setting $\eta \to 0$ when $t \to t_D$ and $\Delta = 0$ when $t \to t_G$, respectively. The variations of t_D and t_G with the coupling ratio α are displayed in Figure 4. It is obvious that the dimerization and the alternating order parameters are decreasing functions of temperature. Since t_D is always larger than t_G, the intermediate temperature state appears, which is called a gapless dimerized state. One can see that the temperature interval between t_D and t_G increases when α increases, and of course its size depends on the strength of the exchange coupling J_0. Owing to the small value of α in crystalline $CuGeO_3$, the gapless SP transition range is very narrow. From the references [6,21], let the parameters for $CuGeO_3$ be $\alpha = 0.1$, $J_0 = 88$ K, and $T_{sp} = 14$ K, the difference between t_D and t_G
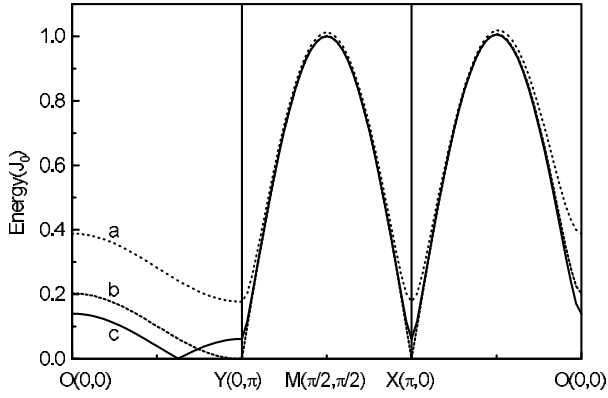
Fig. 3. Energy spectrum ε_k^β in equation (14) in the directions: $O \rightarrow Y$, $Y \rightarrow X$, and $X \rightarrow O$ for $\alpha = 0.1$, (a) $\eta = 0.226$, $P = 0.0565$, (b) $\eta = 0.081$, $P = 0.0202$, and (c) $\eta = 0.0314$, $P = 0.0078$ respectively.
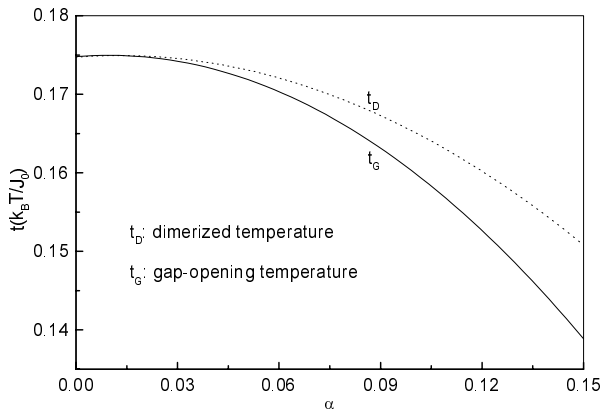

Fig. 4. Dimerization temperature t_D and the gap-opening temperature t_D *versus* α for $K' = 0.708$.

is then approximately 0.6 K which is too small to be observed. This may be a reason why it is difficult to observe experimentally. We suggest that the gapless SP transition phase can be checked by further experiment with large J_0 and α materials.

3 Summary

In conclusion, we have studied the spin-Peierls transition in CuGeO$_3$ based on a quasi-one-dimensional spin-1/2 Heisenberg model by using an extended JWT. The SP phase transition is a two stepped progress: the temperature t_D represents the starting of the gapless dimerized phase, then the energy gap is continuously closed until temperature decreases to t_G at which the opening-gap dimerization happens. It gives strong evidence that the

stepped SP phase depends very crucially on the interchain coupling α. The very small difference between t_D and t_G has not been observed by experiments in the past. It is regarded that in a large inter- and intra-chain coupling magnetic system, the difference may be measured by using X-ray diffraction, nuclear magnetic resonance (NMR), inelastic neutron scattering experiments or other advanced techniques.

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