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The stepped spin–Peierls phase transition in the quasi-one-dimensional spin-1/2 quantum *XY*-model

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Abstract. Using an extension of the Jordan–Wigner transformation (JWT) in two dimensions, the effects of weak interchain coupling on the spin–Peierls (SP) system is studied. The magnetic interaction is considered to be a quantum XY-interaction, and the spin–lattice distortion is treated quasiclassically. On the analogy of the stepped Peierls transition theory, we propose a stepped SP transition theory. We find that, when any finite transverse coupling $J_\perp$ is introduced between nearest chains, the usual SP transition will be modified, and the dimerization and the opening of the gap will no longer occur simultaneously. This leads to the appearance of a gapless SP phase over a certain temperature region.

1. Introduction

The spin–Peierls (SP) transition can be described as a progressive dimerization of spin-1/2 magnetic chains coupled to the three-dimensional (3D) lattice. As the temperature is lowered, the assembly of uniform chains (the U phase) undergoes a second-order phase transition at T_{sp} to a system of dimerized chains (the D phase). The main feature of this state is the appearance of an energy gap between the new ground state—a spin singlet—and the band of the spin-triplet state. Such a transition was first suggested by analogy with the conventional Peierls transition in linear conducting chains. Later, this transition was observed in several organic compounds, such as TTF-Cu(Au)BDT and [MEM$(TCNQ)_2$], where the unpaired electrons are localized in π-orbits of the TTF^+ and $TCNQ^-$ units of the structure [1]. Most recently, Hase, Terasaki, and Uchinokura [2] found that an inorganic compound $CuGeO_3$ in which the $S = 1/2$ spins are localized d electrons of Cu^{2+} ions undergoes a SP transition at 14 K. Since then it has attracted much attention both experimentally and theoretically.

Usually, the theoretical investigations of SP transitions have been performed on the basis of Heisenberg and quantum XY-models. The former includes two successful theories, i.e., those of Pytte [3] and of Cross and Fisher [4]. In these theories, a fermion representation via the Jordan–Wigner transformation (JWT) [5] is used to describe the spin-1/2 chain, and the fermion–phonon interactions are taken into account in the random-phase approximation. The latter [6], i.e., the quantum XY-model mapped also onto the fermion representation via a JWT, though less realistic, presents the advantage of being exactly solvable with respect to the magnetic degrees of freedom. Very recently, Lu *et al* [7] studied the effects of doping

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on SP systems using the unimodular mean-field theory. They concluded that a gapless SP phase occurs at a certain impurity density.

However, the previous theories only consider the intrachain magnetic interaction, while they neglect the interchain coupling, which does exist in real SP materials. It is even more significant for $CuGeO_3$ material. According to Nishi *et al* [8], the ratio of the interchain coupling $J_\perp$ to the intrachain coupling J is about 0.1. This is larger than those in other quasi-one-dimensional systems ($J_\perp/J = 1.7 \times 10^{-2}$ for $CsNiCl_3$ [9] and $J_\perp/J = 4 \times 10^{-4}$ for $Ni(C_2H_8N_2)_2NO_2ClO_4$ [10]). We note that Zhou and Gong [11] considered the effect of the weak interchain coupling on the usual Peierls transition and concluded that the CDW transition and the associated metal–insulator transition no longer occur simultaneously, but now occur at different temperatures. This theory not only can be used to explain the anomalies in some low-dimensional materials such as TaS_3 [12], $NbSe_3$ [13] and $(DMe\text{-}DCNQI)_2Cu$ [14], but also has been verified by new experiments on K-TCNQ [15]. This kind of dimensional effect induced by the weak interchain coupling can be viewed as a general feature of chain-like systems and may have a profound effect on the magnetic properties.

In this paper the SP transition is re-examined by paying special attention to the interchain coupling effect. The interchain coupling is assumed to be weak enough that the 1D feature remains the main one, and sufficiently finite to suppress the thermal fluctuation and to modify the conventional 1D feature. In the following section we employ the extended JWT in two dimensions (2D) to a quasi-1D SP system with a quantum XY-interaction. In section 3, we present the self-consistent equation for the dimerization order parameter and calculate it numerically. Our numerical result seems to support a stepped SP transition. Finally, a brief conclusion is given in section 4.

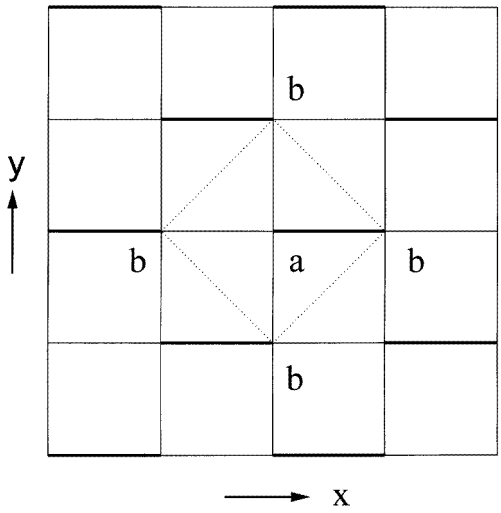

Figure 1. (i) The distribution of the phase factor; each thick line corresponds to a phase factor of 1, and each thin line to a phase factor of $e^{\pm i\pi} = -1$. (ii) The dimerization pattern on a square lattice; each thick and thin line along the x-axis represents stronger coupling $J_+ = J_0(1 + 2\delta u)$ and weaker coupling $J_- = J_0(1 - 2\delta u)$, respectively. Each thin line along the y-axis represents the weak interchain coupling $J_\perp = \alpha J_0$. (iii) The dotted lines indicate the primitive unit cell for both cases (i) and (ii).

2. The Hamiltonian

We begin with the Hamiltonian of a 2D anisotropic quantum XY-model on a square lattice. The Hamiltonian before dimerization is written as

$$H_{xy} = J\sum_{i,j}\{S^x_{i,j}S^x_{i+\hat{\boldsymbol{x}},j} + S^y_{i,j}S^y_{i+\hat{\boldsymbol{x}},j}\} + J_\perp\sum_{i,j}\{S^x_{i,j}S^x_{i,j+\hat{\boldsymbol{y}}} + S^y_{i,j}S^y_{i,j+\hat{\boldsymbol{y}}}\}$$
$$= \frac{J}{2}\sum_{i,j}\{S^-_{i,j}S^+_{i+\hat{\boldsymbol{x}},j} + S^+_{i,j}S^-_{i+\hat{\boldsymbol{x}},j}\} + \frac{J_\perp}{2}\sum_{i,j}\{S^-_{i,j}S^+_{i,j+\hat{\boldsymbol{y}}} + S^+_{i,j}S^-_{i,j+\hat{\boldsymbol{y}}}\} \tag{1}$$

where $\boldsymbol{S}_{i,j}$ is the spin-1/2 operator at $\boldsymbol{r}_{i,j} = i\hat{\boldsymbol{x}} + j\hat{\boldsymbol{y}}$, $\hat{\boldsymbol{x}}$ and $\hat{\boldsymbol{y}}$ 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 in general commute instead of anticommuting. However, one can generalize the conventional JWT to the 2D case by using some approximative schemes [16, 17]. In this paper we adopt the transformation given by Azzouz [17], which is defined on a square lattice as

$$S^-_{i,j} = c_{i,j}\exp\{\mathrm{i}\phi_{i,j}\} \tag{2}$$

$$S^+_{i,j} = \exp\{-\mathrm{i}\phi_{i,j}\}c^\dagger_{i,j} \tag{3}$$

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

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

$$H_{xy} = \frac{J}{2}\sum_{i,j}(\mathrm{e}^{\mathrm{i}\varphi_{i,i+\hat{\boldsymbol{x}};j}}c^\dagger_{i,j}c_{i+\hat{\boldsymbol{x}},j} + \mathrm{HC}) + \frac{J_\perp}{2}\sum_{i,j}(c^\dagger_{i,j}c_{i,j+\hat{\boldsymbol{y}}} + \mathrm{HC}) \tag{5}$$

where $\mathrm{e}^{\mathrm{i}\varphi_{i,i+\hat{\boldsymbol{x}};j}} = \mathrm{e}^{\mathrm{i}[(\phi_{i+\hat{\boldsymbol{x}},j}-\phi_{i,j})-\pi\hat{\boldsymbol{n}}_{i,j}]}$ is a c-number. Equation (5) describes the effective hopping for spinless fermions between nearest sites, of which the hopping amplitudes are $J\mathrm{e}^{\pm\mathrm{i}\varphi_{i,i+\hat{\boldsymbol{x}},j}}$ in the x-direction (parallel to the chains), and $J_\perp$ in the y-direction (perpendicular to the chains). There are many different configurations for the phase factor $\mathrm{e}^{\mathrm{i}\varphi_{i,i+\hat{\boldsymbol{x}};j}}$ depending on the spin configuration. Thus, we can get a state similar to the known mean-field solution of the in-phase flux state [16] by choosing the phase factor as in figure 1, in which each thick line corresponds to a phase factor of 1, and each thin line to a phase factor of $\mathrm{e}^{\pm\mathrm{i}\pi}$. This distribution ensures that each of the elementary plaquettes encloses a net flux of half-quanta.

Since the undistorted state has been discussed thoroughly by several authors [16, 17], we now turn to the dimerized state (dimerized along the x-axis). Among many dimerization patterns on a square lattice, we choose one according to an elastic neutron scattering study of $CuGeO_3$ (see figure 1 of reference [18]). We note that this dimerization pattern coincides with the distribution of the phase factor of the in-phase flux state, i.e., each thick and thin line along the x-axis represents 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 interchain coupling $J_\perp = \alpha J_0$. Here J_0 is the exchange integral along the x-axis before dimerization and $\delta = -\mathrm{d}J^x_{i,j}/\mathrm{d}u$. The spin–lattice distortion u is treated quasiclassically as Su, Schrieffer, and Heeger (SSH) [19] did for linear conducting chains. Following the

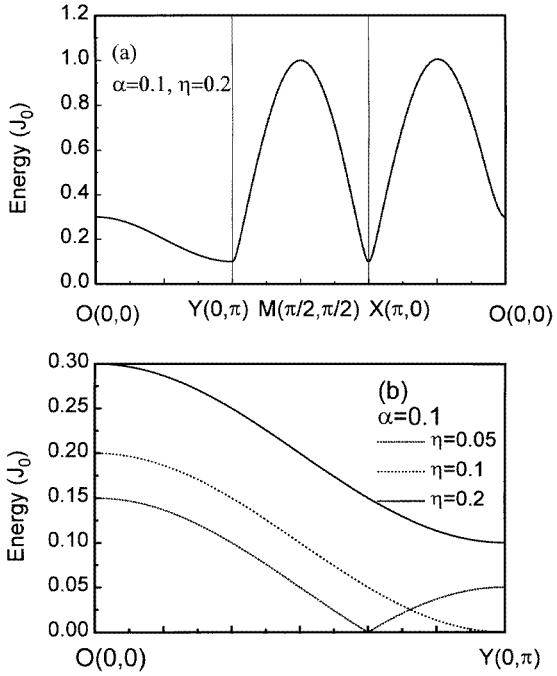

Figure 2. (a) The energy spectrum ϵ_k^β in equation (12) in the directions O $\rightarrow$ Y, Y $\rightarrow$ X, and X $\rightarrow$ O for $\alpha = 0.1$, and $\eta = 0.2$. (b) The energy spectrum ϵ_k^β in equation (12) in the direction O $\rightarrow$ Y for $\alpha = 0.1$, $\eta = 0.05$, 0.1 and 0.2 respectively.

notation and the distribution of the coupling in figure 1, the dimerized Hamiltonian of the quasi-1D spin-1/2 quantum XY-model can be expressed in terms of the spinless fermion operators, a and b, of the two sublattices A and B, respectively:

$$H = \sum_{r_{i,j}\in A}\left\{-\frac{J_-}{2}(b^\dagger_{i-\hat{x},j}a_{i,j} + \text{HC}) + \frac{J_+}{2}(a^\dagger_{i,j}b_{i+\hat{x},j} + \text{HC}) + \frac{\alpha J_0}{2}(b^\dagger_{i,j-\hat{y}}a_{i,j} + b^\dagger_{i,j+\hat{y}}a_{i,j} + \text{HC})\right\} + \frac{1}{2}\sum_{r_{i,j}\in A} K[(u_{i,j} - u_{i-\hat{x},j})^2 + (u_{i+\hat{x},j} - u_{i,j})^2] \tag{6}$$

where $u_{i,j} = (-1)^{i+j}u$ is the spin–lattice distortion, and K is the elastic constant. For $CuGeO_3$, the neutron scattering study [18, 20] shows the dimerization of Cu–Cu pairs along the x-axis with an interatomic distance of 2.930 Å, and the separation between dimers is 2.955 Å while the alternative coupling is about 120 K and 110 K. This allows us to assume that u is negligible as compared with the lattice spacing a, and $2J_0\,\delta u$ is not negligible as compared with J_0. Thus, the Hamiltonian (6) can be written in momentum space as

$$H = \frac{1}{2}\sum_{BZ'}\{(-J_-\mathrm{e}^{\mathrm{i}k_x a} + J_+\mathrm{e}^{-\mathrm{i}k_x a} + 2\alpha J_0\cos k_y a)a^\dagger_k b_k + \text{HC}\} + 2NKu^2 \tag{7}$$

where BZ′ is the magnetic Brillouin zone. This Hamiltonian (7) can be easily diagonalized as

$$H = \left\{ \sum_{BZ'} E_k^{\alpha} \alpha_k^{\dagger} \alpha_k + E_k^{\beta} \beta_k^{\dagger} \beta_k \right\} + 2NKu^2 \tag{8}$$

with the energy spectra

$$E_k^{\alpha} = -\sqrt{J_0^2 \sin^2 k_x a + (2\,\delta u \cos k_x a + \alpha J_0 \cos k_y a)^2} \tag{9}$$

$$E_k^{\beta} = \sqrt{J_0^2 \sin^2 k_x a + (2\,\delta u \cos k_x a + \alpha J_0 \cos k_y a)^2}. \tag{10}$$

It is evident that, in the pure 1D limit ($\alpha = 0$), our energy spectra E_k^{α} and E_k^{β} coincide with those obtained in previous SP theory [6]; and in the undistorted case, equations (9) and (10) are reduced to the spectra of the in-phase flux state [16, 17].

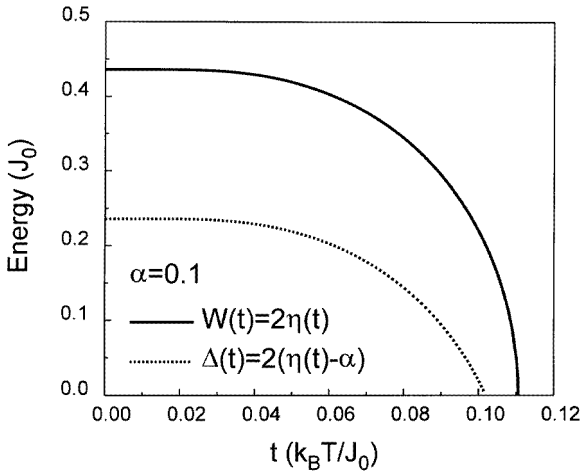

Figure 3. The temperature (t-) dependences of the dimerization order parameter $W(t) = 2\eta(t)$ and the energy gap $\Delta(t) = 2(\eta(t) - \alpha)$ for $\alpha = 0.1$ and $K' = 0.6107$.

3. The order parameter and the stepped SP transition

For convenience, we define a set of reduced parameters: $J_0 = 1$, $a = 1$, the reduced temperature $t = k_B T / J_0$, the dimerization order 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

$$\epsilon_k^{\alpha} = -\sqrt{\sin^2 k_x + (\eta \cos k_x + \alpha \cos k_y)^2} \tag{11}$$

$$\epsilon_k^{\beta} = \sqrt{\sin^2 k_x + (\eta \cos k_x + \alpha \cos k_y)^2}. \tag{12}$$

The spectrum ϵ_k^{β} for $\alpha = 0.1$ and $\eta = 0.2$ is shown in figure 2(a). In the directions O $\rightarrow$ Y and X $\rightarrow$ O, it is in good agreement with the recent experimental results on $CuGeO_3$

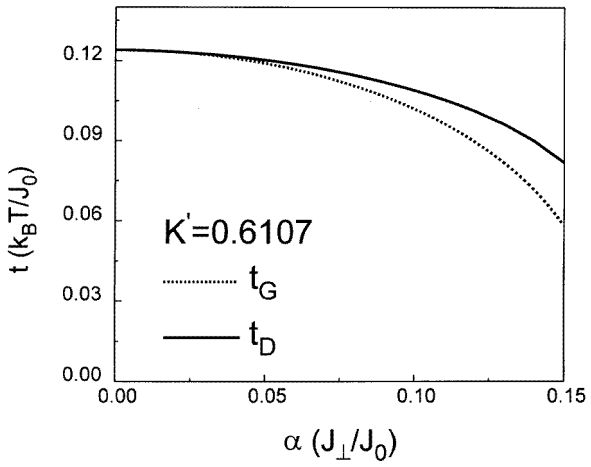

Figure 4. The α-dependences of the dimerization temperature (t_D) and the gap-opening temperature (t_G) for $K' = 0.6107$.

obtained using inelastic neutron scattering (see figure 5 and figure 8 of reference [20]; the experimental result for the direction Y $\rightarrow$ X is not available in this reference). From the spectra ϵ_k^α and ϵ_k^β (figure 2(b)), we can see that the weak interchain coupling modifies the spectra, and the energy gap becomes

$$\Delta = \min(\epsilon_k^\beta) - \max(\epsilon_k^\alpha) = 2(\eta - \alpha). \tag{13}$$

It turns out that, when $\eta(t=0) < \alpha \neq 0$, $\Delta = 0$ at any temperature, while dimerization is permissible; when $\eta(t=0) > \alpha$, the dimerization and the opening of the gap will occur at different temperatures due to the different temperature dependences of $\Delta(t)$ and $\eta(t)$. To show these results in detail, we now turn to the numerical calculation. The reduced free energy per unit cell can be derived by using the diagonalized Hamiltonian (8):

$$\begin{aligned} f = -\frac{2}{J_0 N} k_B T \ln Z &= -\frac{2t}{N} \ln(\mathrm{Tr}\, \mathrm{e}^{-H/k_B T}) \\ &= -\frac{2t}{N}\left\{\sum_{BZ'} \ln 2\cosh\frac{E_k^\alpha}{2t} + \ln 2\cosh\frac{E_k^\beta}{2t}\right\} + K'\eta^2 \\ &= -t\int\int_{BZ'} \frac{\mathrm{d}k_x\ \mathrm{d}k_y}{2\pi^2}\left\{\ln 2\cosh\frac{\epsilon_k^\alpha}{2t} + \ln 2\cosh\frac{\epsilon_k^\beta}{2t}\right\} + K'\eta^2. \end{aligned} \tag{14}$$

From the condition $\partial f/\partial\eta = 0$, we obtain the self-consistent equation for the dimerization order parameter $\eta(t)$:

$$K' = \int\int_{BZ'} \frac{\mathrm{d}k_x\ \mathrm{d}k_y}{8\pi^2}\ \frac{(\eta\cos k_x + \alpha\cos k_y)\cos k_x}{\eta\sqrt{\sin^2 k_x + (\eta\cos k_x + \alpha\cos k_y)^2}}\left\{\tanh\frac{\epsilon_k^\beta}{2t} - \tanh\frac{\epsilon_k^\alpha}{2t}\right\}. \tag{15}$$

Then, we can calculate the dimerization temperature t_D and the gap-opening temperature t_G numerically by setting $\eta \rightarrow 0$ when $t \rightarrow t_D$ and $\eta \rightarrow \alpha$ when $t \rightarrow t_G$, respectively. The numerical results are presented in figure 3 and figure 4.

Since the dimerization order parameter $\eta(t)$ is a decreasing function of temperature (figure 3), t_G is always less than t_D; thus there exists an intermediate-temperature region in which the dimerization occurs while the gap does not open as can be seen from figure 3 and

figure 4. This indicates that a gapless SP phase appears. So, the SP transition in this spin-1/2 quasi-1D quantum XY-system is essentially a two-step process: the system is driven into the gapless SP phase at the first critical temperature t_D; and the gap remains closed until the temperature is lowered to the second critical temperature t_G at which the gap opens and the gapped SP phase forms. We can also see that t_D and t_G depend crucially upon the coupling ratio α. Only in the pure 1D case ($\alpha = 0$) can the two critical temperatures t_D and t_G coincide. Because the coupling ratio α in a real system is small, the gapless SP phase is very narrow. For $\alpha = 0.1$, the difference between t_D and t_G is estimated to be $0.01J_0$, which is very small compared with $t_D = 0.11J_0$ (see figure 3). If we use the parameters for $CuGeO_3$ ($\alpha = 0.1$, $J_0 = 125$ K, $T_{sp} = 14$ K) [8, 21], the difference between t_D and t_G is estimated to be about 1.3 K. This may be why this gapless SP phase has not been observed experimentally in the past. Anyway, our prediction of a gapless SP phase can be checked by further experiments. t_D can be measured in x-ray diffraction or NMR experiments, and t_G can be measured using inelastic neutron scattering. And magnetic materials with large J_0 and large α are recommended for detecting this gapless SP phase.

4. Conclusion

In this paper we have studied the effect of the weak interchain coupling on the quasi-1D spin-1/2 quantum XY-model by using an extended JWT in two dimensions. Our calculations reveal that a new phase, i.e., the gapless SP phase, may occur between the conventional uniform phase and the gapped SP phase. This indicates that the weak interchain coupling can also lead to the appearance of a gapless SP phase, as the doping does [7].

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