Mean-field theory of the spin-Peierls systems: Application to CuGeO$_3$

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A mean-field theory of the spin-Peierls systems based on the two dimensional dimerized Heisenberg model is proposed by introducing an alternating bond order parameter. Improvements with respect to previous mean-field results are found in the one-dimensional limit for the ground state and the gap energies. In two-dimensions, the analysis of the competition between the antiferromagnetic long range order and the spin-Peiers ordering is given as a function of the coupling constants. We show that the lowest energy gap to be observed does not have a singlet-triplet character in agreement with the low temperature thermodynamic properties of CuGeO$_3$.

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The recent discovery of a spin-Peierls (SP) transition in the inorganic compound CuGeO$_3$ prompts renewed interest for this kind of structural instability. Evidence for a non-magnetic transition has been exemplified in several ways. The lattice distortion has been well established by X-ray and elastic neutron experiments. The magnetic susceptibility decreases exponentially showing a gap in the spin excitations. This is also confirmed by heat capacity measurements which present a thermally activated component below the SP critical temperature. As shown by Oseroff et al., another interesting feature is the close proximity between the SP and the antiferromagnetic (AF) ground state for doped CuGeO$_3$ samples.

For the theoretical description of the SP ordered state, the alternating Heisenberg model has been intensively studied both numerically and analytically. However in the latter, mean-field like decoupling of the Heisenberg interaction in the quasi-fermion representation were essentially restricted to the single chain problem. In the present work, we introduce a new decoupling for the alternating bond order parameter which not only improves 1D mean-field results for the ground state energy and the excitation gap but also allows to treat the 2D situation, namely the effect of interchain Heisenberg exchange interaction. This turns out to be an important ingredient for the interpaly between the AF and the SP states.

We start the analysis with the 2D alternating Heisenberg model

$$H = J \sum_{i,j} S_{2i,j} \cdot S_{2i+1,j} + J' \sum_{i,j} S_{2i+1,j} \cdot S_{2i+2,j} + J' \sum_{i,j} S_{i,2j} \cdot S_{i,2j+1} + \frac{1}{2} \sum_{i,j} C_{i,j}^2 \cdot S_{i,2j+1} \cdot S_{i,2j+1}$$

where $J > 0 (J' > 0)$ and $J' > 0 (J'' > 0)$ are the intrachain (interchain) AF exchange couplings. We then follow the mean-field approach given in refs. [12] and [13]. In such a treatment, the spin Hamiltonian is transformed by means of the generalized Jordan-Wigner (JW) transformation

$$S_{i,j}^- = c_{i,j} e^{i(\sum_{\ell=1}^{j-1} n_{\ell,j} + \sum_{\ell=0}^{j-1} n_{i,\ell})}$$

$$S_{i,j}^+ = c_{i,j}^* e^{-i/2}$$

where $n_{i,j}$ is the order parameter. The Hamiltonian is then written in Fourier space by taking into account the bipartite character of the lattice, namely

$$H = \frac{1}{2} \sum_{k} \{ M_{k}^A \phi_{k}^A - M_{k}^B \phi_{k}^B + e(k) \phi_{k}^{A} \phi_{k}^{B} + e^*(k) \phi_{k}^{B} \}$$

where $u_k = e^{i\alpha_k/2} \cos \beta_k$ and $v_k = e^{i\beta_k/2} \sin \beta_k$. $\alpha_k$ and $\beta_k$ are given by

$$\tan \alpha_k = \frac{(J_1 - J_2) \sin k_x + (J_{11} - J_{12}) \sin k_y}{(J_1 + J_2) \cos k_x + (J_{11} + J_{12}) \cos k_y}$$

and

$$\tan (2\beta_k) = \pm (2M)_{-1}$$

where

$$\frac{[\cos k_x (J_{11} + J_{12}) \cos k_y]^2 + [(J_1 - J_2) \sin k_x + (J_{11} - J_{12}) \sin k_y]^2}{2}$$

where $J_1 = J(1+2Q)$, $J_2 = J'(1+2Q')$, $J_{11} = J_{12} (1+2P')$ and $J_{12}$ = $J_{12} (1+2P')$. As for the dispersion relation, it is given by

$$E_{\pm}(k) = \pm \frac{1}{2} (M^2 + |e(k)|^2)^{1/2}$$

where (±) refers to upper and lower band. We have introduced the order parameters $m = 2 \langle S_z \rangle$, $Q = |\langle c_{2i,j} c_{2i+1,j}^2 \rangle|$, $Q' = |\langle c_{2i+1,j} c_{2i+2,j}^2 \rangle|$, $P = |\langle c_{i,j} c_{i+1,j}^2 \rangle|$. 

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and \( P' = |c_{z,2j+1}c_{z,2j+2}^\dagger | \) for the staggered magnetization, intra- and interchain bond amplitudes respectively.

Their equilibrium values are obtained from the minimization of the total free energy leading to a set of mean field equations which can be solved exactly.

The ground state wave function corresponds to the case where the lower band is filled:

\[
|\Phi_{GS}| = \prod_k d^\dagger_{k}\ket{0}.
\]

It is formed by the pairs of fermions \( (c^\dagger_k, c^\dagger_k) \) with the weights \( v_k \) and \(-u_k\) respectively which correspond from \( \mathbb{Z}_2 \) to pairs of spins denoted \( (\uparrow, \downarrow)_k \) in reciprocal space. The ground state is a singlet since \( \langle w^\dagger \rangle \) the compressibility in the JW representation. Its evaluation for each member of the particle-hole pair is given by

\[
S = \sum_{\kappa} \text{phase factors in (2) have been ignored in or-}
\]

\[
\text{The field equations which can be solved exactly.}
\]

\[
\text{by the pairs of fermions \( (c^\dagger_k, c^\dagger_k) \) with the weights \( v_k \) and \(-u_k\) respectiv}
\]

and the compressibility in the JW representation. Its evaluation is straightforward and the result is given by

\[
\chi(T) = -\frac{1}{2} g^2 \mu^2 \sum_k \frac{\partial n(E_k(k))}{\partial E_k(k)}
\]

\[
\sim \frac{1}{2} g^2 \mu^2 \mathcal{D}(E_{GS}^{SP}) e^{-\beta E_{GS}^{SP}} \quad (\beta E_{GS}^{SP} \gg 1)
\]

where \( n[x] = (e^{\beta x} + 1)^{-1} \) is the Landé factor and \( \mathcal{D}(E_{GS}^{SP}) \) is the density of states at the energy gap. Therefore the lowest energy gap to be observed in experiments (e.g. in CuGeO\(_3\)) like magnetic susceptibility and specific heat of the condensed SP state is characterized by the above particle-hole (singlet) character. Furthermore, in contrast to the critical temperature, the amplitude of the zero temperature gap is not predicted to change when a magnetic field is applied in agreement with specific heat and acoustic measurements performed under low field.

When both possibilities of SP and AF long range order are considered (Eq. 18), the numerical solution of the mean field equations leads to the phase diagram of Fig. 1 for \( J'/J, J_{zz}/J \) at a fixed \( J'_z \) value. Whenever \( m \neq 0 \), the rotational invariance is broken and the system is in the AF state while the line boundary is determined when \( m \) vanishes. As for the SP phase, it is defined by \( m = 0 \) and an energy gap solely due to dimerization. In the insert of Fig. 1 the magnetization is displayed as a function of \( J'/J \) for \( J_{zz} = J'_z = 1.1J \). Therefore whenever the dimerization becomes sufficiently small, the SP ordered state is no longer stable and a magnetic ordering is favored; an increase of the interchain exchanges also favors the magnetic ordering. This is consistent with the situation found in real systems like quasi-1D organic materials where the application of hydrostatic pressure is well known to promote such a crossover.

Focussing now on the ground state energy \( E_{GS} = \langle \Phi_{GS}|H|\Phi_{GS} \rangle \) in the \( m = 0 \) SP phase, one can compare in Figure 2 the present mean-field approach with the previous results obtained by Bulaevskii in the 1D limit. Thus the choice of an altered order parameter \( (Q \neq Q') \) gives rise to a better estimation of the ground state energy. In the uniform Heisenberg limit of the model, \( J = J' \), the dispersion relation \( E_+ (k) = (1 + 2Q) |\sin k| \) becomes gapless with \( 1 + 2Q \approx 1.63 \) in fair agreement with the exact result.

As far as the gap is concerned in this 1D limit (insert of the Figure 2), the present approach leads to

\[
E_{GS}^{SP} = E_0 + C \mid 1 - J'/J \mid^\alpha,
\]

with \( C \approx .8J \) and the exponent \( \alpha \approx .71 \) which is close to the Cross and Fisher value \((\alpha = 2/3)\) and exact diagonalization \((\alpha = .79 \pm .06)\). The present calculation however predicts a finite jump \( E_0 \approx .19J \) for the energy gap once \( J = J' \) is non zero; a result not yet confirmed by an exact numerical calculation probably due to finite size effects as \( J - J' \rightarrow 0 \).

When the effect of interchain exchange coupling is included (Figure 3), one can extract in the region where the
SP phase is stable (for \( J_\perp / J < .45 \), the AF phase is dominant for all \(| J - J' |\)) the universal value \( \alpha \simeq .66 \) for the exponent, while the constant \( E_0 \) decreases monotonously with \( J_\perp \) and \( C \simeq .81 J \). As a 2D mean-field result, this value of \( \alpha \) is closer to the one obtained by Cross and Fisher.\(^\text{17}\)

In summary, we have proposed a mean-field theory of the 2D dimerized Heisenberg model with magnetic and alternating bond order parameters for the description of the ordered spin-Peierls state and its competition with the antiferromagnetic order. In the 1D limit, the ground state energy and the particle-hole excitation gap profiles with dimerization show marked improvements with respect to previous mean-field results. As far as the excitation gap is concerned, particle-hole excitations should dominate the thermodynamics at low temperature and this, consistently with recent measurements on specific heat and magnetic susceptibillity for the CuGeO\(_3\) material.

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FIG. 1. The phase diagram (\( J'/J, J_\perp/J \)) is drawn for \( J'_\perp = .08 J \). In the insert, the magnetization \( m \) is reported as a function of \( J'/J \) for \( J'_\perp = J_\perp = .1 J \).

FIG. 2. The ground state energy as a function of \( | J' - J | \) in the present mean-field approximation compared to the Bul‘evskii’s results (B). In the insert, the same comparison for 1D SP gap.

FIG. 3. The energy gap \( E_0^{SP} \) as a function of \( | J - J' | \) for \( J_\perp = .15 J \) (curve 1) and \( J_\perp = .1 J \) (curve 2). The curve 3 gives the energy gap which would be obtained in the absence of antiferromagnetism \( (m = 0) \) for \( J_\perp = .15 J \). In the insert, the magnetization is reported for \( J_\perp = .15 J \) (curve 1) and \( J_\perp = .1 J \) (curve 2).