

## Doping Effect on Spin-Peierls Instability

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We study the effects of doping on spin-Peierls (SP) systems using the unimodular mean-field theory. The impurity spins affect the singlet valence bond field and renormalize the magnetic excitations. The SP transition temperature and the energy gap of magnetic excitations are reduced by factors  $\sim n_i$  and  $\sim n_i^{2/3}$ , respectively, with  $n_i$  as the impurity density. At a certain value of  $n_i$ , a gapless SP phase occurs, and the interaction between impurities becomes RKKY-like. The recently observed reduction of SP transition temperature upon doping and occurrence of a spin glass phase is interpreted using the proposed theory.

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The theoretically predicted spin-Peierls (SP) state with alternating bond length has been observed, so far only in a few quasi-one-dimensional (1D) organic compounds with antiferromagnetic (AF) interactions [1]. Very recently, it was found in an inorganic compound  $\text{CuGeO}_3$  [2]. Moreover, a drastic reduction of SP transition temperature  $T_c$  was observed upon Zn doping and a spin glass (SG) phase appeared in the doping range  $0.02 < n_i < 0.08$  [3]. In this Letter we propose a theory of SP transition in doped systems explaining these findings and making further predictions on a *gapless SP state* to be checked by experiments.

The SP transition is driven by the interaction between the (1D) spin-1/2 chains and the three-dimensional (3D) lattice, which makes a mean-field (MF) approach available due to its suppression of fluctuations. Below the SP transition a uniform AF chain is deformed into an alternating AF chain with a singlet ground state and a magnetic gap [4]. Up to now there are two successful theories of SP transition, i.e., that of Pytte [5] and of Cross and Fisher [6]. In these theories, a fermion representation via the Jordan-Wigner transformation (JWT) is used to describe the spin-1/2 chain, and the fermion-phonon interactions are taken into account in the random phase approximation. However, it is difficult to investigate effects of impurity doping upon the SP systems within this approach due to nonlocal features of JWT.

Anderson [7] has proposed the resonant-valence-bond model to describe two-dimensional spin-1/2 AF systems. Later, Arovas and Girvin [8] put forward a unimodular mean-field theory (UMFT) to decouple the quartic term in an  $S = 1/2$  uniform AF chain by a unimodular Hubbard-Stratonovich transformation. In this scheme the quantum spins are represented by fermion operators, while the uniform valence bond (VB) formed by two nearest neighbor  $S = 1/2$  spins as a singlet is fixed to unity, so the constraint of one fermion per site is satisfied on average. Therefore, the fluctuations of the VB field are drastically suppressed, and the results obtained are sat-

isfactory.

In this Letter we employ the UMFT to study the SP system, and consider the SP order parameter, the spin-lattice distortion  $u$  quasiclassically as Su, Schrieffer, and Heeger (SSH) [9] did in linear conducting chains. All the known results of the SP transition obtained earlier [5,6,10] can be readily derived in our approach. The advantage of this scheme is the easiness to deal with the doping effects. In particular, it provides a clear picture about the impurity effects on the SP system, analogous to the doped  $S = 1$  linear-chain AF with a Haldane gap [11,12]. To our knowledge, this is the first paper to deal theoretically with doped SP systems.

Suppose a SP system is doped randomly by impurities with different spins which disturb the VB structure of the SP system, namely, the VB field is modified due to the impurity spin-flip terms, and the wave function of the magnetic excitation is renormalized by the impurity scatterings. We find the SP  $T_c$  is reduced by a factor  $\sim n_i$  with  $n_i$  as the impurity density, whereas the decrease of the energy gap is proportional to  $n_i^{2/3}$  and it collapses at some value of  $n_i$ , but the system is still in the SP phase, i.e., the spin-lattice dimerization remains. Thus we predict the existence of a *gapless SP state* to be checked by direct experiments. This is rather similar to superconductors doped with paramagnetic impurities which reduce the energy gap and eventually give rise to gapless superconductivity [13].

The Hamiltonian describing an impurity-doped SP system is [12,14]

$$\begin{aligned} H &= H_0 + H^{\text{im}}, \\ H_0 &= \sum_l J(l, l+1) \mathbf{S}_l \cdot \mathbf{S}_{l+1} + \sum_l 2K u^2, \\ H^{\text{im}} &= \sum_{\langle l\alpha \rangle} \mathbf{A}_{\alpha l} \cdot \mathbf{S}_l, \\ \mathbf{A}_{\alpha l} &= g(\alpha, l) \mathbf{S}_{\alpha}^{\text{im}} - J(\alpha, l) \mathbf{S}_{\alpha}, \quad J(l, l+1) > 0, \end{aligned} \quad (1)$$

where  $\mathbf{S}^{\text{im}}$  is the impurity spin,  $g$  is the coupling between impurity and host spins,  $\alpha$  denotes impurity site,  $\langle l\alpha \rangle$

means summation over all impurity sites with  $l = \alpha \pm 1$ ,  $u$  is the lattice distortion, and  $K$  is the elastic constant.

We consider a random distribution of impurities along the spin chain. Suppose no correlations exist between  $\mathbf{S}_\alpha^{\text{im}}$  and  $\mathbf{S}_\alpha$  if  $\mathbf{S}_\alpha^{\text{im}} \neq \mathbf{S}_\alpha$ , then it is appropriate to assume that  $\overline{\mathbf{S}^{\text{im}}} = 0$ ,  $\overline{\mathbf{S}_\theta^{\text{im}}} = \frac{1}{3}S^{\text{im}}(S^{\text{im}} + 1)$  (where  $\theta$  means any direction),  $\overline{\mathbf{A}_{\alpha l}^2} = g(\alpha, l)^2 S^{\text{im}}(S^{\text{im}} + 1) + J(\alpha, l)^2 S(S + 1)$ , if averages are taken over impurity's random distribution. Of course,  $\overline{\mathbf{A}_{\alpha l}^2} = [g(\alpha, l) - J(\alpha, l)]^2 S(S + 1)$ , if  $\mathbf{S}_\alpha^{\text{im}} = \mathbf{S}_\alpha$ . Since the ion mass is much bigger than the electron mass, the kinetic energy of the lattice can be neglected, so we can consider the lattice distortion quasiclassically. Moreover, the impurity spins are also considered as quasiclassical quantities in our treatment (implications of the quantum nature for impurity spins will be mentioned later). The host  $S = 1/2$  quantum spins are represented by fermion operators as [8],  $S^+ = f_\uparrow^\dagger f_\downarrow$ ,  $S^- = f_\downarrow^\dagger f_\uparrow$ ,  $S^z = \frac{1}{2}(f_\uparrow^\dagger f_\uparrow - f_\downarrow^\dagger f_\downarrow)$  with a constraint  $f_\uparrow^\dagger f_\uparrow + f_\downarrow^\dagger f_\downarrow = 1$ .

We assume that the spin lattice is divided into two

$$H = 2N(J_0 S^2 + 2Ku^2 + \lambda) + \frac{1}{2} \sum_k \Psi^\dagger(k) (|\Delta_k| \Omega_5 + \lambda \Omega_4) \Psi(k) + \frac{1}{2N} \sum_{k, k'} \Psi^\dagger(k) [\mathbf{S}(k - k') \cdot \boldsymbol{\Omega}] \Psi(k'), \quad (2)$$

where

$$\Psi^\dagger(k) = (f_{k\uparrow}^{\dagger A} \ f_{k\downarrow}^{\dagger A} \ f_{k\downarrow}^A \ f_{k\uparrow}^A \ f_{-k\uparrow}^{\dagger B} \ f_{-k\downarrow}^{\dagger B} \ f_{-k\downarrow}^B \ f_{-k\uparrow}^B),$$

$\Psi(k)$  is its Hermitian conjugate,  $\lambda$  is the Lagrangian multiplier for the constraint  $f_\uparrow^\dagger f_\uparrow + f_\downarrow^\dagger f_\downarrow = 1$ , and

$$\Delta_k = -\sqrt{2}(J_A e^{-ik(1+2u)} + J_B e^{ik(1-2u)}),$$

$$\mathbf{S}(k - k') = - \sum_{\alpha \in A \text{ or } \alpha \in B} e^{i(k-k')(\alpha+1)} (J_B e^{-i2(k-k')} + J_A) \mathbf{S}$$

with  $J_B = (J_0 - 2\alpha u)/2$  and  $J_A = (J_0 + 2\alpha u)/2$ , while  $\Omega_i = \sigma_i \otimes \sigma_3 \otimes I$  ( $i = 1, 2$ ),  $\Omega_3 = \sigma_3 \otimes I \otimes I$ ,  $\Omega_4 = I \otimes \sigma_3 \otimes I$ ,  $\Omega_5 = \sigma_3 \otimes \sigma_1 \otimes \sigma_1$ , and  $\boldsymbol{\Omega} = (\Omega_1, \Omega_2, \Omega_3)$ , with  $I$  and  $\sigma_1, \sigma_2, \sigma_3$  as  $2 \times 2$  unit and Pauli matrices. The lattice constant is set to unity, and the spin chain has  $2N$  sites. Note that we have performed the gauge transformation of Read and Newns [15] to absorb the phase factor of the VB, and make the VB be unity in Eq. (2) to suppress its fluctuations (for details see Ref. [8]).

First consider the undoped case to check our formalism. The Hamiltonian can be exactly diagonalized, and the MF free energy per site can be easily derived, from which in the limit  $k_B T \ll \sqrt{2}J_0$ , the SP  $T_c$  is given as,  $k_B T_c = 1.13\sqrt{2}J_0 e^{-1/\lambda_0}$ , with  $\lambda_0 = 4\alpha^2/\pi K\sqrt{2}J_0$ . At zero temperature, for small  $u$  one finds  $\alpha^2 u^2/J_0^2 = 4/(e^{2+2/\lambda_0} + 8 - 24/\lambda_0)$ . If a magnetic field  $h$  less than a critical value is applied, the SP state is disturbed, but the dimerization still remains [10]. For small  $h$ , it is easy to obtain  $T_{ch} = T_{c0}[1 - 0.32(\mu_B h/k_B T_{c0})^2]$ , where  $T_{c0}$  is determined in zero magnetic field,  $\mu_B$  is Bohr magneton. This magnetic field dependence is a special feature of the

sublattices  $A$  and  $B$  along the chain, and the exchange integral is expanded in terms of the small lattice distortion  $u$  as  $J(l, l + 1) = J_0 + 2\alpha u$ , for  $l \in A$  sublattice, and  $J(l, l + 1) = J_0 - 2\alpha u$ , for  $l \in B$  sublattice, where  $\alpha = -dJ(l, l + 1)/dl$  and  $J_0$  is the exchange integral for a uniform chain.

As a result of the spin-flip scatterings, it is convenient to use the eight-component generalization of the Nambu formalism to treat the Hamiltonian (1). In Hase *et al.*'s experiment [3] nonmagnetic impurities were employed. For simplicity we will also discuss the nonmagnetic doping, although the formalism itself is very general. Of course, if the spin chain is strictly 1D, doping by nonmagnetic impurities will sever the spin chain. However, a real system is only quasi-1D, so the interchain coupling as well as the superexchange between the next nearest neighbors will carry over the spin correlation effects. Since impurities are randomly distributed, the doping of two sublattices should be equivalent after averaging. In UMFT, the Hamiltonian (1) can be rewritten in the momentum space as

SP transition distinguishing it from ordinary structural transitions.

Now we define an  $8 \times 8$  Matsubara matrix Green function of the SP system as  $G(k, k'; \tau) = -(T_\tau \Psi(k, \tau) \Psi^\dagger(k', 0)) = \beta^{-1} \sum_n G(k, k'; i\omega_n) e^{-i\omega_n \tau}$ , where  $\Psi(k, \tau)$  is the imaginary time Heisenberg representation of  $\Psi(k)$ ,  $T_\tau$  denotes the imaginary time order,  $\langle \dots \rangle$  indicates the thermal average, and  $\omega_n = (2n + 1)\pi/\beta$  for the fermionic model. The impurity scattering breaks the translational invariance which is recovered after averaging over the random impurity configurations. Then the Matsubara Green function will take the form  $\delta_{kk'} G(k, i\omega_n)$  satisfying the Dyson equation,  $G^{-1}(k, i\omega_n) = G_0^{-1}(k, i\omega_n) - \Sigma(k, i\omega_n)$ , where  $\Sigma(k, i\omega_n)$  is the self-energy term, while the bare Matsubara Green function  $G_0(k, i\omega_n)$  is given from Eq. (2) by  $G_0^{-1}(k, i\omega_n) = i\omega_n \Omega_0 - |\Delta_k| \Omega_5 - \lambda \Omega_4$ , with  $\Omega_0$  as a  $8 \times 8$  unit matrix. Using the standard technique for resumming diagrams [13], we find

$$\Sigma(k, i\omega_n) = \left(\frac{1}{N}\right)^2 \sum_{k_1} [\mathbf{S}(k_1 - k) \cdot \boldsymbol{\Omega}] \times G(k_1, i\omega_n) [\mathbf{S}(k - k_1) \cdot \boldsymbol{\Omega}]. \quad (3)$$

Considering the properties of the matrices  $\boldsymbol{\Omega}$ ,  $\Omega_4$ , and  $\Omega_5$ , we make an ansatz

$$G^{-1}(k, z) = \bar{Z}(k, z) z \Omega_0 - \bar{Z}(k, z) |\bar{\Delta}_k(z)| \Omega_5 - \bar{Z}(k, z) \bar{\lambda}(k, z) \Omega_4, \quad (4)$$

where  $z$  is the frequency in the complex plane,  $\bar{Z}(k, z)$  is the wave function renormalization factor,  $|\bar{\Delta}_k(z)|$  and

$\bar{\lambda}(k, z)$  are the renormalized parameters for the VB field and the Lagrangian  $\lambda$  multiplier, respectively.

Substituting Eqs. (4) and (3) into the Dyson equation yields a set of integral equations,

$$\begin{aligned} \bar{Z}(k, z)z &= z - \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \frac{dk_1}{\pi} K(k, k_1; z) \bar{Z}(k_1, z)z, & \bar{Z}(k, z)\bar{\lambda}(k, z) &= \lambda + \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \frac{dk_1}{\pi} K(k, k_1; z) \bar{Z}(k_1, z)\bar{\lambda}(k_1, z), \\ \bar{Z}(k, z)|\bar{\Delta}_k(z)| &= |\Delta_k| + \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \frac{dk_1}{\pi} K(k, k_1; z) \bar{Z}(k_1, z)|\bar{\Delta}_{k_1}(z)|, \\ K(k, k_1; z) &= \frac{n_i S(S+1)[J_A^2 + J_B^2 + 2J_A J_B \cos 2(k - k_1)]}{\bar{Z}(k_1, z)^2 z^2 - \bar{Z}(k_1, z)^2 |\bar{\Delta}_{k_1}(z)|^2 - \bar{Z}(k_1, z)^2 \bar{\lambda}(k_1, z)^2}, \end{aligned} \quad (5)$$

with  $n_i$  as the impurity density. Meanwhile, the constraint becomes

$$\frac{2}{\beta} \sum_n \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \frac{dk}{\pi} \frac{\bar{Z}(k, i\omega_n)(i\omega_n) + \bar{Z}(k, i\omega_n)\bar{\lambda}(k, i\omega_n)}{\bar{Z}(k, i\omega_n)^2 (i\omega_n)^2 - \bar{Z}(k, i\omega_n)^2 |\bar{\Delta}_k(i\omega_n)|^2 - \bar{Z}(k, i\omega_n)^2 \bar{\lambda}(k, i\omega_n)^2} e^{i\omega_n 0^+} = 1. \quad (6)$$

Now Eqs. (5) and (6) form a closed set of equations to determine the parameters  $\bar{Z}(k, z)$ ,  $|\bar{\Delta}_k(z)|$ ,  $\bar{\lambda}(k, z)$ , and  $\lambda$ , and the Green function  $G(k, z)$ . In the doped case the impurity spins affect the distribution of the spin density along the chain, so the constraint can no longer be automatically satisfied.

By the Feynman theorem, the free energy  $F$  of the doped SP system is  $F = F_0 + \int_0^1 \frac{\langle \rho H^{\text{im}} \rangle_\rho}{\rho} d\rho$ , where  $\langle \dots \rangle_\rho$  means the thermal average for the Hamiltonian  $H_\rho = H_0 + \rho H^{\text{im}}$ , and  $F_0$  is calculated for  $\rho = 0$ . Suppose  $n_i$  is very small, so we keep the free energy  $f$  per site only up to the first order of  $n_i$ , and obtain  $T_c$  in the doped case. If a weak magnetic field  $h$  is also employed, the crossing terms of  $h$  and  $n_i$  will emerge in the free energy. However, we may reasonably omit them for small  $n_i$  and  $h$ . Thus the magnetic field and the impurities will independently change the properties of the SP system. It is readily shown that the SP  $T_c$  is

$$T_c = T_{c0} \left[ 1 - 0.14n_i \left( \frac{\sqrt{2}J_0}{k_B T_{c0}} \right)^2 - 0.32 \left( \frac{\mu_B h}{k_B T_{c0}} \right)^2 \right], \quad (7)$$

where  $T_{c0}$  is given in zero magnetic field and without doping. This equation is very important for verifying the SP instability.

The fermion density of states is given by

$$\mathcal{N}(z) = \frac{1}{8} \int \frac{dk}{\pi} \left( \text{Tr} \frac{\text{Im} G(k, z)}{-\pi} \right).$$

For small  $n_i$ , we may also expand this equation up to the first order of  $n_i$ . After a somewhat lengthy calculation (see [16]), we find a threshold frequency

$$\omega_c = 2\sqrt{2}\alpha u \left( 1 - \frac{q^{\frac{2}{3}}}{16\alpha^2 u^2} + \frac{p}{24\alpha^2 u^2} \right), \quad (8)$$

with  $q = 12\pi n_i \frac{\alpha^2 u^2 J_0^2}{\sqrt{2}J_A J_B}$ ,  $p = n_i(10.5\alpha^2 u^2 - 0.375J_0^2)$ . For  $\omega < \omega_c$ , the density of states  $\mathcal{N}(\omega)$  is zero. A physical magnetic excitation has spin  $S = 1$ . In our approach it is made up of two  $S = 1/2$  fermions. In the weakly doped case we may ignore the correlation effect between the

two fermions induced by scattering at the same impurity. Thus, the energy gap of the doped SP system is  $E_G = 2\omega_c$ . This dependence upon  $n_i$  is similar to that of the Haldane energy gap upon doping [12].

In the experimental papers [2,3], Hase *et al.* adopt the exchange integral  $J_0 = 88$  K ( $k_B = 1$ ) according to Bonner and Fisher's work [17], and the SP transition is observed around 14 K. Thus at  $T = 0$  K the SP order parameter  $u$  and the energy gap  $E_{G_0}$  in the absence of impurities are given by  $\alpha u/J_0 = 0.076$ ,  $E_{G_0} = 4\sqrt{2}\alpha u = 38$  K. Using these parameters as input, we find  $T_c = T_{c0}(1 - 11.1n_i)$ , which is in good agreement with the experimental result of Hase *et al.* It is thus inferred that for  $n_i$  close to 10%, the  $T_c$  vanishes and the SP state disappears. In the weakly doped case one can replace  $4\sqrt{2}\alpha u$  approximately by  $E_{G_0}$  and obtain from Eq. (8),  $E_G = E_{G_0}(1 - 7.9n_i^{2/3} - 10.4n_i)$ , from which we find that the gap vanishes at  $n_i \sim 2.8\%$ .

Now we can give the following physical picture. The impurity doping greatly affects the properties of the SP system, especially it has a more drastic impact upon the density of states than the SP  $T_c$  itself. When the impurity concentration  $n_i$  increases, the energy gap  $E_G$  reduces faster than the SP  $T_c$ . At some value of  $n_i$  (about 0.03 for the nonmagnetic doping), the energy gap first collapses, but the spin-lattice dimerization still remains. The spin lattice does not become uniform until another value of  $n_i$  (about 0.10). We call the state in between a *gapless SP phase*. This interpretation is consistent with the experimental observations of [3], because the SP transition was determined there by a sudden drop of the magnetic susceptibility which shows the presence of a spin gap. We would, therefore, prefer to interpret the critical concentration  $n_c = 0.03$  as the collapse of the spin gap, rather than the SP state itself. The weaker singularity in susceptibility than  $T^{-1}$  is also consistent with the reduced density of states in the pseudogap. Of course, a direct check of our prediction would be the measurement of the gap itself by neutron scattering and determination of the VB alternation by x-ray diffraction or nutation NMR experiments. The dimerization should depend both on the

doping and the strength of the applied magnetic field.

The SG behavior observed in [3] was interpreted there mainly due to the interchain coupling and other frustration effects. In our view, it is more related to the gapless spin excitation spectrum in this doping range of the SP state. In fact, from model (2) we see that the correlation between impurities is mediated by fermions. In the usual SP phase a magnetic excitation has a gap  $E_G$ , so the interaction between two impurities induced by the magnetic excitations should be proportional to  $e^{-L/\xi}$ , where  $L$  is the distance between the two impurities,  $\xi$  is the correlation length and is proportional to  $E_G^{-1}$ . It is obvious that such short range correlation does not favor the SG state. To the contrary, in the gapless SP phase  $\xi$  is infinite, and the magnetic excitations are gapless. The correlations between impurities should be of a power law. We may approximately evaluate the second order effective interaction between the impurities corresponding to the Feynman vacuum polarization graph as [16]

$$H_{\text{eff}} \propto \sum'_{\alpha, \gamma} \frac{4\pi J_0^2 \cos \pi(|R_\alpha - R_\gamma|)}{|R_\alpha - R_\gamma|} \mathbf{S}_\alpha \cdot \mathbf{S}_\gamma,$$

where  $R_\alpha$  is the  $\alpha$ th impurity position, and  $\sum'$  means  $\alpha \neq \gamma$ . This correlation is RKKY-like.

It appears to us that the combination of long-range correlations between impurities and spin-lattice distortion leading to frustrations are mainly responsible for the SG behavior. Of course, further studies are needed to clarify this issue. It is worthwhile to mention that the gapless spin excitation spectrum in some doped  $S = 1$  Haldane AF [12] should also lead to long-range correlations among impurity spins, and possibly, a SG behavior as well.

Up to now, we have considered only the scattering effects of doped impurities. However, multiple scattering of spin excitations on impurities may give rise to bound states in the gap, as in superconductors with magnetic impurities [18] and  $S = 1/2$  state in Haldane gap AF chain [19,12]. Moreover, the quantum fluctuations of impurities may also lead to some observable effects [20]. These issues will be discussed in future publications.

To summarize we have developed a theory to consider the doping effect on the SP state. The results obtained on the reduction of SP transition temperature and spin gap upon doping agree with experiments. The prediction of a gapless SP state can be checked by further experiments.

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