On the Incommensurate Phase of Pure and Doped Spin-Peierls System $CuGeO_3$

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Phases and phase transitions in pure and doped spin-Peierls system $CuGeO_3$ are considered on the basis of a Landau-theory. In particular we discuss the critical behaviour, the soliton width and the low temperature specific heat of the incommensurate phase. We show, that dilution leads always to the destruction of long range order in this phase, which is replaced by an algebraic decay of correlations if the disorder is weak.

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The spin-Peierls (SP) transition is the classic instability of one dimensional quantum spin-half antiferromagnetic chains due to the coupling of the spins with the lattice. A rigid Heisenberg chain has a nonmagnetic uniform ground state with a gapless fermionic excitation spectrum [1]. This can be seen most easily by using the Jordan-Wigner transformation, which maps the spins onto (strongly) interacting pseudo-fermions [2]. Due to the coupling to the lattice the system can lower its energy by the standard Peierls mechanism: lattice distortions freeze in at a wave vector $2k_F$ which leads simultaneously to the opening of a gap at the Fermi-level in the fermionic spectrum such that the energies of all occupied fermionic states decrease [3]. In zero magnetic field the free-fermion band is half-filled with Fermi-wave vector $k_F = \frac{\pi}{2a}$, which corresponds to a dimerization of the chain. A non-zero magnetic field lowers the Fermi-level [4], but Umklapp processes still favor the distortion at π/a until a critical field strength H_I is reached, at which a transition to an incommensurate (I) phase with modulation vector $|2k_F - q_s|$ sets in. In the I-phase a new (empty) band appears in the middle of the gap of the fermionic spectrum. Thus, spin excitations still exhibit a gap which is however smaller than the gap of the dimerized (D) phase. The above picture follows from theories obtained for free or weakly interacting pseudo fermions, in which phonon dynamics were essentially ignored [5,6]. There, the SP transition is the result of the freezing of a (classical) phonon mode due to further downwards renormalization of the phonon frequency by the spin-phonon interaction [5,6]. This scenario is supported by the experimental data of organic SP systems [7]. However, it does not seem to apply in all respects to the transition found recently in the inorganic SP substance $CuGeO_3$ [8]. Though not devoid of controversy, there is now a wealth of well-accepted results for $CuGeO_3$, which shows two SP transitions [9–13,15,16]. The SP transition from a disordered, uniform (U) to a D phase at 14.3 K in zero field is shifted slightly to lower T if the field increases until a Lifshitz point at $T \approx 11.3$ K and $H \approx 12.5$ T is reached, where the transition to an I phase sets in. Some experimental results which are not explained by the existing theories, are: (i) no soft phonon has been observed so far [8,9], (ii) a (Peierls) gap in the D phase is observed in low temperature specific heat measurements, but not in the I phase, for which a Debye–like T^3-law has been found with an amplitude much larger than the background (lattice) contribution [10], (iii) solitons, which are supposed to produce the modulation in the I phase are broad [11] in comparison to the sharp Sine-Gordon like solitons predicted by mean-field like calculations [5], (iv) already a small amount of doping leads to a strong reduction of the SP temperature T_{SP} [16] and a drastic suppression of the anomalies at the UI transition [11,17].

Since the phonon energies are always large compared with the magnetic ones, the applicability of the adiabatic approximations has been questioned [18]. Khomskii et al. [19] developed a soliton picture of the SP transition in $CuGeO_3$, which resembles somewhat structural order-disorder transitions [20]. No soft mode phonon is expected, but the SP transition corresponds to deconfinement of solitons, which are bound to pairs below T_{SP}. These solitons are simultaneously magnetic and structural excitations: they carry spin 1/2 and are domain walls between the two groundstates of the dimerized lattice.

It is the aim of the present paper to explain the properties (ii)–(iv) by a pure phenomenological approach, which avoids delicate approximations in the coupled spin phonon system: The T^3-law of the specific heat in the I phase is explained quantitatively by phason fluctuations. It is argued that broad solitons are fingerprints of the type II lock-in transition which occurs in SP systems like $CuGeO_3$. Finally, we show that dilution leads to complete destruction of long range order in the I phase.

Incommensurate phases are classified according to the existence of an inversion symmetry for the structural transition in question [21]. In case there is an inversion symmetry for the Hamiltonian, as for $CuGeO_3$, first derivatives of the order parameter (Lifshitz invariants) do not exist. Indeed, for $CuGeO_3$ the uniform high temperature orthorhombic structure, space group

$Pbmm$, changes below $T_{\rm SP}$ to a dimerized structure, space group $Bbcm$, with distortion wave vector $(\frac{1}{2}\frac{1}{2}0)$ (established from X-ray and neutron diffraction experiments [9]). Standard group theoretic arguments based on the symmetries and the invariant group of the distortion vector in the Brillouin zone [22] show, that the transition is described by four non-equivalent, one dimensional irreducible representations [23]. It is very likely, that only one of these four representations corresponds to the primary order parameter, which is real and can be considered to be proportional to the displacement of the copper ions. The other three may occur as secondary order parameters. In fact, neutron scattering data indicates that at least two normal modes are necessary to explain the displacement pattern of the D phase [9]. A simple transformation of reversing the displacements on one sublattice helps us in getting an ordered state with zero wavevector for the D phase. We take this transformed and coarse grained displacements to be proportional to the order parameter field $\psi(\mathbf{x})$. In general, $\psi(\mathbf{x})$ will also include contributions from the magnetic degrees of freedom.

The Landau Hamiltonian is that of an anisotropic Ising model [24,21] $\mathcal{H}\{\psi\} = \int d^d\mathbf{x}\, h\{\psi(\mathbf{x})\}$, where the Hamiltonian density is (for $i = x, y, z$)

$$h\{\psi\} = \frac{r}{2}\psi^2 + \sum\nolimits_i \frac{c_i}{2}(\partial_i\psi)^2 + \frac{u}{4}\psi^4 + \\ + \frac{d}{4}(\partial_z^2\psi)^2 + \frac{w}{6}\psi^6 + \frac{e}{2}\psi^2\,(\partial_z\psi)^2 \qquad (1)$$

Terms involving $\partial_z\psi\,\partial_z^3\psi$, though not shown explicitly, may also appear [21]. We have included higher order terms in order to stabilize the system for the case when one of the c_i coefficients (here c_z) or u becomes negative for sufficiently strong magnetic field. A negative c_i signals the transition to the I phase. The parameters r and c_i are taken as analytic functions of T and H with $r = r_0\,(T - T_{\rm SP}(H))$, and $c_z(T, H) = c_0\,(H_{\rm I}(T) - H)$. Microscopic treatments [5] and experiments suggest [10], that u also decreases considerably with increasing magnetic field.

A *mean field analysis* (MFA) of the phase diagram requires treating the Hamiltonian Eq. (1) as a free energy minimized with respect to ψ. Let us first assume, that u and d remain positive everywhere in the H–T–plane. Then, we can ignore the last two terms in (1). If $c_i > 0$ for all i, the minimum of the free energy occurs for wave vector $\mathbf{k} = 0$, while a nonzero $\mathbf{k}$ vector is possible if $c_z < 0$. The mean field phase boundaries are given for UD: $r = 0$, $c_z > 0$, UI: $r = c_z^2/2d$ and DI: $r = \frac{1}{4}(\sqrt{3/2} - 1)^{-1}c_z^2/d$ [24]. The spontaneous wave vector in the I phase is given by $q_s^2 = -c_z/d$. The DI transition is *first order* while the other two are second order, in agreement with experiments.

Fluctuations do make subtle changes in the phase diagram but the overall features remain the same. The critical behavior of the UD transition is Ising–like but the UI transition is XY–like. This difference originates from the fact, that in the I phase the order parameter condenses at $\mathbf{k} = \pm\mathbf{q}_s$ and has consequently two components $\mathbf{\Delta}(\mathbf{x}) = (\Delta_1(\mathbf{x}), \Delta_2(\mathbf{x}))$. These are related to $\psi(\mathbf{x})$ by $\psi(\mathbf{x}) = \sqrt{2}\,[\Delta_1(\mathbf{x})\cos(\mathbf{q}_s\mathbf{x}) + \Delta_2(\mathbf{x})\sin(\mathbf{q}_s\mathbf{x})]$. In the I phase, the Landau functional thus can be written as

$$h\{\mathbf{\Delta}\} = \tfrac{1}{2}(r - c_z^2/2d)\mathbf{\Delta}^2 + 2|c_z|(\partial_z\mathbf{\Delta})^2 \\ + c_x(\partial_x\mathbf{\Delta})^2 + c_y(\partial_y\mathbf{\Delta})^2 + \tfrac{3}{2}u(\mathbf{\Delta}^2)^2 \qquad (2)$$

Since the number of degrees of freedom of the system cannot change when going from the D to the I phase, it is clear that Eq. (2) is valid only for fluctuations of $\mathbf{\Delta}(\mathbf{x})$ with wavelength long compared to q_s^{-1}, i.e. as long as we are away from the Lifshitz point.

If one approaches the ordered phase along the line $c_z(T, H) = 0$, one observes so called *Lifshitz critical behaviour*, which follows from a change of the dispersion relation to $A_{\mathbf{k}} = r + c_x k_x^2 + c_y k_y^2 + dk_z^4$. Note that at the Lifshitz critical point the conventional hyperscaling is changed to $\nu_\| + (d-1)\nu_\perp = 2-\alpha$ where $\nu_\| = \nu_\perp/2 = 0.31$ are the correlation length exponents parallel and perpendicular to the z-direction [24]. Approaching the D or I phase, respectively, from the U phase on a line parallel to that given by $c_z(T, H) = 0$, at first Lifshitz type critical behaviour will be observed before the region of Ising– or XY–type critical, respectively, behaviour is asymptotically reached.

Considering the DI transition, fluctuation effects are expected to be less important, because it is first order in MFA. A refined MFA has been worked out by Bruce, Cowley and Murray [21] for this case, who found that in the I phase the order parameter can be described by a multiplane-wave Ansatz $\psi(\mathbf{x}) = \sum a_m \cos(mq_z z)$ with $m = 1, 3, 5...$, which is rapidly converging. For example, the ratio $|a_3/a_1| \approx 0.035$ close to the transition [21]. In this sense the system shows *broad* domain walls. Also in this refined theory the transition remains first order.

Above we assumed u to be *positive* even for large field values. In the opposite case, the transition to the U phase might become first order. Some mean-field theories [5] predict very special relations between the coefficients of the Landau-expansion, i.e.

$$u/c_z = \text{const.}\,, \quad w = 3du^2/4c_z^2\,, \quad e = 5du/2c_z\,. \qquad (3)$$

If these are fulfilled, the DI transition may become *continuous*, at least close to the Lifshitz point [25]. Indeed, for this very particular relation of the coefficients of the Landau-expansion (1), the ground state solutions are the Jacobian elliptic functions $\psi_s(z) \equiv \sin(\phi_s(z)/2) = \psi_0\,\mathrm{sn}(z/k\xi_s, k)$, where ξ_s is a bare correlation length (expressed by c_z, d and r) and k is the modulus of this function [5,26]. Note, that $\phi_s(z)$, which is related to the spin-density, obeys the Sine-Gordon equation. In this case solitons are *sharp* in the sense that the separation of domain walls diverges by approaching the D phase. However, from a symmetry point of view, which we adopt here, we do not see a deeper reason, why the relations (3) should be fulfilled in general by an *exact* microscopic theory. In fact, these relations were obtained using the adiabatic approximation. Consequently one has to expect, that in general the w- and e-terms in (1) *do* exist, but violate the relations (3). These terms will change the modulation amplitude ratio $|a_3/a_1|$ to larger values, but without reaching the sharp soliton limit. Thus the DI transition is expected to remain first order, as found also experimentally for $CuGeO_3$ [8,11].

Although the validity of Landau-theory is essentially restricted to the region close to the transition, one should expect that it can be used to understand at least qualitatively the low temperature specific heat data. For this purpose, we have to determine the low-lying excitations of the ordered structure. These can be found by adding the kinetic energy term $\int \mathrm{d}^d x \frac{\varrho}{2}\dot{\psi}^2(\mathbf{x})$ to the GL-Hamiltonian, where the mass density ρ will have contributions both from the magnetic and the lattice degrees of freedom. We will further assume, that ψ obeys Bose-statistics. Using the saddle point approximation to determine the equilibrium value of ψ, one obtains in the D phase $\omega^2(\mathbf{k}) = \frac{1}{\rho}(2|r| + c_i k_i^2)$ for the frequency of the harmonic excitations of the order parameter field. In the D phase, were the order parameter is real, we identify $E_g = \hbar(2|r|/\rho)^{1/2}$ with the gap which is found in the low-T specific heat. In the I phase in addition to the massive amplitude mode a gapless *phason* mode with frequency $\omega^2(\mathbf{k}) = \frac{1}{\rho}\left(c_x k_x^2 + c_y k_y^2 + 2|c_z| k_z^2\right)$ appears [21], which will dominate the specific heat

$$C_{\text{phason}} \approx \frac{\sqrt{2}\pi^2}{15} k_B \left(\frac{k_B T}{E_g \overline{\xi}_0}\right)^3 \equiv \beta_{\text{phason}} T^3. \qquad (4)$$

Here we have introduced $\overline{\xi}_0 = (\xi_{0x}\xi_{0y}\xi_{0z})^{1/3}$ where $\xi_{0i}^2 = c_i/r_0 T_0$ and used $T \approx T_0/2$ to express ρ by E_g. Thus, the phason mode delivers a T^3–contribution to the low–temperature specific heat, in addition to that from acoustic phonons [27].

Next we extend our analysis to the *quenched disordered case*, e. g. random substitutions of Cu by Zn or Ni and/or Ge by Si in $CuGeO_3$. Such substitutions change the various interactions locally but do not break the symmetry of the displacements in favor of a particular dimerization. Therefore, the effects of these random substitutions can be modeled by randomness in the coefficients of the original Landau Hamiltonian without any symmetry breaking term. Little reflection shows, that the main effect will come from a randomness $\delta r(\mathbf{x})$ in r [28]. In the D phase, this leads to a decrease of T_{SP}, as was shown microscopically by Khomskii et al. [19]. Moreover, the critical behaviour will be changed to that of the diluted Ising model [29].

The effect of disorder is even more severe in the I phase. This can be seen easily by rewriting $\psi(\mathbf{x})$ as $\psi(\mathbf{x}) = \sqrt{2}\Delta(\mathbf{x})\cos(\mathbf{q}_s\mathbf{x} + \theta(\mathbf{x}))$. With $\delta r(\mathbf{x}) = \kappa\sum_i \delta(\mathbf{x} - \mathbf{x}_i)$ the disorder term can now be written as

$$\frac{\kappa}{2}\sum_i \Delta^2(\mathbf{x}_i)\, \cos\left[2\left(\theta(\mathbf{x}_i) + \mathbf{q}_s\mathbf{x}_i\right)\right] \qquad (5)$$

The random impurity positions $\mathbf{x}_i$ lead to a random phase $\alpha_i \equiv \alpha(\mathbf{x}_i) = 2\mathbf{q}_s\mathbf{x}_i \pmod{2\pi}$ which is equally distributed between 0 and 2π. It is well known that such a random anisotropy term destroys the translational long range order of the I phase [30]. However, the phase–phase correlation function diverges only logarithmically [31] $\overline{\langle(\theta(\mathbf{x}) - \theta(\mathbf{0}))^2\rangle} = \frac{\pi^2}{18}\ln(x/L_L)$. Here the overbar denotes the disorder average. The Larkin–length L_L is related to the strength of the disorder, a rough estimate is

$$L_L \approx 2\pi^3\left[\bar{\xi}_0^2/(d\ln T_{\text{SP}}/dn_{\text{imp}})\right]^2 n_{\text{imp}}^{-1} \qquad (6)$$

where n_{imp} denotes the concentration of the impurities. Because of the logarithmic divergence of the phase fluctuations, there is however *quasi–long range order* of the order parameter correlation function

$$\overline{\langle\mathbf{\Delta}(\mathbf{x})\mathbf{\Delta}(\mathbf{0})\rangle} \approx \left(\bar{\xi}_0\sqrt{\textstyle\sum_i (x_i/\xi_{0,i})^2}/L_L\right)^{-\pi^2/36} \qquad (7)$$

Despite of the loss of true long range order, the system will however still show Bragg peaks of finite width, as follows from the Fourier transform of (7). In deriving these results we have neglected vortex-ring excitations. It has been argued recently, that these can indeed be neglected for sufficiently weak disorder and low temperatures [32]. At elevated temperatures or larger dilution their condensation triggers the transition to the disordered phase. The type of this transition is presently unknown.

We briefly apply the results obtained so far to $CuGeO_3$. Fixing the $T = 0$ value of the order parameter at $\psi_0 = 1$, we have $r_0 T_0 = u_0$. From the mean–field jump of the specific heat $\Delta C_{\text{MFA}} = r_0^2 T_0/2u = u_0/2T_0 \approx 22.7\frac{\text{mJ}}{\text{K}\,\text{cm}^3}$ in zero magnetic field [10], we get $u_0 = 650\frac{\text{mJ}}{\text{cm}^3}$ which gives the correct size of the critical region. Since ΔC_{MFA} decreases for increasing field and is reduced approximately by a factor 4.6 when reaching the Lifshitz point, u is reduced correspondingly to about $u_L = 112\frac{\text{mJ}}{\text{cm}^3}$, but still positive. Defining the Ginzburg critical region $\tau_G \equiv |T_G - T_0|/T_0$ as the region, in which the first fluctuation correction to the specific heat becomes larger than ΔC_{MFA}, this gives for zero field, $\xi_{0,x} = 0.12\,\text{nm}$, $\xi_{0,y} = 0.36\,\text{nm}$ and $\xi_{0,z} = 0.69\,\text{nm}$ [14], with geometric mean $\overline{\xi}_0 = 0.31\,\text{nm}$, $\tau_{G,I} \approx (k_B T_0/8\pi u\overline{\xi}_0^3)^2 \approx 0.16$ – larger ΔC_{MFA} diminishes τ_G correspondingly. For the XY-transition far from the Lifshitz point we get $\tau_{G,XY} \approx 0.32$ at a magnetic field where

$\xi_z(H, T = T_{\rm SP}) \approx \xi_{0,z}$. At the Lifshitz point the critical region is given by $\tau_{G,L} \approx \tau_G^{2/3}(\xi_{0,z}u_0/\sqrt{2}\zeta u_L)^{4/3} \approx 0.7$ where $\zeta_0 = (d/2r_0T_0)^{1/4} \approx 1.2\,\mathrm{nm}$. The critical exponent β changes from $\beta_I \simeq 0.325$ for $H < H_L$ to $\beta_L \simeq 0.15..0.18$ for $H \approx H_L$ and then to $\beta_{\rm XY} \simeq 0.346$ for $H > H_L$, in agreement with the experimental observation [13].

From the low–temperature specific heat in the D phase one finds $E_g \approx 23\,\mathrm{K}$ [10,14], which gives with Eq. (4) for the phason specific heat $\beta_{\rm phason} \approx 1.3\,\frac{\rm mJ}{\rm K^4 mol}$ in the I phase, which has to be compared with the experimental value of $1.4\,\frac{\rm mJ}{\rm K^4 mol}$ [10]. This good agreement is possibly to some degree accidental, since the magnetic field dependences of the various parameters have not been taken into account carefully. But at least the order of magnitude should be right.

For the Larkin length we obtain with $x = n_{\rm imp}v_{\rm uc}/2$ for the concentration of the Zn-atoms ($v_{\rm uc}$ denotes the volume per unit cell) and assuming a linear dependence of $T_{\rm SP}(x)$ on x with $d\ln T_{\rm SP}(x)/dx \approx 14$ [19], for $x \approx 0.04 : L_L \approx 1.2\,\mathrm{nm}$ and for $x \approx 0.07 : L_L \approx 0.7\,\mathrm{nm}$. The data of Kiryukhin et al. [11] was fitted with an exponential decay of correlations with an anisotropic correlation length ξ of order 10 nm. It would be interesting to check, whether their data can also be fitted by a power law (7).

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