Ordered phases in spin-Peierls systems

Götz S. Uhrig

Institut für Theoretische Physik, Zülpicher Straße 77, Universität zu Köln, D-50937 Köln, Germany

Abstract

The microscopic description of spin-Peierls substances is discussed. Particular attention is paid to the ordered (dimerised and incommensurably modulated) phases. Important points are the adiabatic and the antiadiabatic approach, generic soliton forms, elastic and magnetic interchain couplings. The wealth and the accuracy of experimental information collected for the first inorganic spin-Peierls substance CuGeO$_3$ motivates this work.

Keywords: non-adiabaticity; soliton form; local renormalisation; interchain coupling

The coupling of 1D magnetic and 3D elastic degrees of freedom in spin-Peierls systems leads to magnetically driven lattice distortions at low $T$ (see e.g. [1,2]). Without magnetic field the spin chains dimerise (D phase). For sufficiently large magnetic field or by defects the homogeneous dimerisation is modulated. Solitons occur: zeros of the dimerisation at which $S = 1/2$ spinons are localised [3,4]. The high-magnetic field, incommensurate (I) phase is characterised by an array of these solitons.

Since we deal with coupled magnetic and elastic degrees of freedom the microscopic Hamiltonian is $H = H_S + H_{SB} + H_B$. The purely magnetic part is given by

$$H_S = J \sum_{i,j} (S_{i,j}S_{i+1,j} + \alpha S_{i,j}S_{i+2,j})$$

(1)

where $i$ is the site index within a given chain $j$. The frustrating next nearest neighbour coupling $\alpha J$ is included since there is evidence that this term is important in CuGeO$_3$ [5-7]. The phonons are treated as free bosons

$$H_B = \sum_q \omega(q) b_q^\dagger b_q$$

(2)

The Peierls phenomenon stems from the interaction $H_{SB}$. Since the atomic displacements are small we assume that they influence the magnetic exchange in linear order

$$H_{SB} = \sum_q A_q (b_q^{+} b_{-q})$$

(3)

with

$$A_q = \sum_k g(q,k) S_k S_{-k}$$

(4)

Unfortunately, this Hamiltonian is too complicated. So one argues that the 1D magnetic subsystem strongly favours a distortion at wave vector $\pi$ which is underlined by the dimerisation below the transition temperature $T_{SP}$ and by the diverging dimer-dimer correlation at $\pi$ for $T \to 0$. Hence those phonons matter most which are in the vicinity of the actual lattice distortion at $q^d$ for $T < T_{SP}$. It appears permissible to take their energy
\( \omega(q^d) \) as the energy of all the phonons neglecting the phonon dispersion. This does not imply that the 3D character is lost since in generic lattices with basis even the local phonons are extended objects (for the polarization vectors of CuGeO3 see [8–10]). This fact leads to the important elastic coupling between adjacent chains so that the spin-Peierls transition can take place at \( T > 0 \). A purely 1D model would not display a phase transition at \( H \) in real space reads

The operators \( A_{i,j} \) far from the point indexed by \( g \), e.g. \( A_{i,j} = g(S_{i,j} - S_{i-1,j} + f \sum_{x} (S_{i+1,j} \pm S_{i,j+1} - S_{i,j} \pm S_{i-1,j+1})) \) [11]. The parameter \( f (|f| < 1) \) measures how much a phonon on chain \( j \) influences also its adjacent chains \( j \pm 1 \).

Now two routes can be followed. The first, more traditional, route consists in treating the phonons adiabatically, i.e. the distortions are considered as static

\[
\langle b_{i,j} \rangle =: \delta_{i,j} J/(2g)
\]  

in the ordered phases. Correspondingly, there is no influence of \( H_{SB} \) above \( T_{SP} \), see e.g. Ref. [12]. The phonon propagators are renormalised in RPA by the magnetic response in the dimer-dimer channel [13]. The transition is signalled by the vanishing of the renormalised phonon frequency, the so-called phonon softening. This description applies if the phonons are the slow degrees of freedom [13] which are renormalised by the fast magnetic degrees of freedom. An obvious condition for the validity of this scenario is \( \omega < J \).

It was shown by numeric investigation that the more restrictive condition \( \omega < \Delta \) applies where \( \Delta \) is the gap in the D phase [14]. The gap \( \Delta \) is a measure for the effect of the spin-phonon coupling, i.e. \( \Delta \) indicates up to which energy the magnetic and the phononic degrees of freedom are decisively altered by the coupling. Hence the phonon cannot become soft if \( \omega > \Delta \) holds. This view, suggested in particular for the spin-Peierls substance CuGeO3 [11], is corroborated by field theoretic results [15].

The second route consists in the antiadiabatic approach which considers the spin system as slow and the phononic system as fast. Then it is reasonable to eliminate the phonons in favour of an effective spin model. Many works took actually this approach [16,17] in leading order in \( J/\omega \). Couplings dependent on \( T \) arise in the next-leading order [11]. The transition into the ordered phase does not occur due to the softening of a phonon but due to the tendency of the effective spin model towards dimerisation. Considering a single chain phonon-induced frustration above its critical value \( \alpha_{\text{eff}} > \alpha_c \approx 0.241167(5) \) [18] drives the dimerisation [11,19,17]. Hence a finite spin-phonon interaction is necessary to achieve the spontaneous symmetry breaking [20–22].

In Sect. 1 we compare the adiabatic and the antiadiabatic route to the spin-Peierls transition. In Sect. 2 the I phase will be discussed in detail. A summary concludes this article.

1. Mean-field, nonadiabaticity and phasons

Most adiabatic approaches use the Hamiltonian

\[
H = J \sum_i \left[ (1 + \delta_i)S_{i+1}S_i + \alpha S_{i+2}S_i + \frac{K}{2} \delta_i^2 \right]
\]

with some spring constant \( K \) (see e.g. [23,24]); the chain index has been dropped. The elastic interchain coupling does not appear since its main effect can be absorbed in the 1D model as long as the modulations on neighbouring chains occur in phase. An important exception to this rule are solitons induced by impurities [25,26,4]. The local distortions \( \delta_i \) in Eq. (6) are subject to the constraint

\[
\sum_i \delta_i = 0
\]

The ground state energy \( \langle H \rangle \) is minimised with respect to the \( \delta_i \) leading to

\[
K \delta_i = -\langle S_{i+1}S_i \rangle + \langle S_{i+1}S_i \rangle
\]

where the overline stands for the sample average to ensure the constraint.

For \( S_{\text{tot}} = 0 \), i.e. in the D phase, the solution to the minimisation of Eq. (6) reads \( \delta_i = (-1)^i \delta_0 \).
where the amplitude $\delta_0$ is proportional to the expectation value of the dimerisation operator $H_D = \sum_i (-1)^i S_{i+1} S_i$, i.e. $\delta_0 = \langle H_D \rangle / K$ (see e.g. [27]). For finite magnetisation solitons occur. A generic solution for $S_z = 1$ is displayed in Fig. 1 [3].

What does one obtain in the antiadiabatic limit? For simplicity we assume $A_{i,j} = g(\hat{S}_{i+1,j} \hat{S}_{i,j} : + f \sum_{\pm} : \hat{S}_{i+1,j,\pm 1} \hat{S}_{i,j,\pm 1} :)$ where the colons stand for the operator reduced by its expectation value ($: X := X - \langle X \rangle$). This is to prevent “trivial” renormalisation of the bare coupling constants. Applying a unitary transformation eliminating $H_{SB}$ one obtains in the two leading orders additional terms in the spin Hamiltonian $\Delta H_X = O(g^2/\omega)$ and $\Delta H_Y = O(g^2/J/\omega^2)$ [11]

$$\begin{align*}
\Delta H_X &= -\frac{1}{\omega} \sum_{i,j} A_{i,j}^+ A_{i,j} \\
\Delta H_Y &= \frac{1}{2\omega^2} \coth \left(\frac{\omega}{2T}\right) \sum_{i,j} [A_{i,j}^+ L A_{i,j}] ,
\end{align*}$$

(8) (9)

where $L$ is the Liouville operator, i.e. the commutation with $H_S$. In $\Delta H_Y$ the interchain terms in $A_{i,j}$ do not generate qualitatively new terms. In $\Delta H_X$, however, the interchain terms lead to terms

$$H_{\text{int}} = -\frac{2g^2 f}{\omega} \sum_{i,j} : \hat{S}_{i+1,j} \hat{S}_{i,j} : : \hat{S}_{i+1,j+1} \hat{S}_{i,j+1} :.$$

(10)

coupling adjacent chains. It is obvious that via $H_{\text{int}}$ the dimerisation on one chain favours dimerisation on the adjacent chain. So in the antiadiabatic description the part (10) is responsible for the phase transition at $T > 0$ since it creates the coherence of the dimerisation pattern throughout the whole sample.

It is often argued that the adiabatic phonon description works very well, see e.g. [12,10,15]. The pieces of evidence, Ginzburg criterion, susceptibility and entropy fits, do indeed favour a mean-field description. They do not, however, require an adiabatic phonon description. In view of $\omega \gg \Delta$ in CuGeO$_3$ [11], we advocate the antiadiabatic phonon description. This is combined with a chain-mean field treatment (CMF) since $TS_P/J \approx 0.1$ is small.

Within each chain an effective Hamiltonian as derived previously [11] is used. Between the chains the contribution (10) is treated in mean-field. Let us abbreviate

$$\delta_i := -\frac{2g^2 f}{\omega J} \sum_{\pm} \langle : \hat{S}_{i+1,j,\pm 1} \hat{S}_{i,j,\pm 1} : \rangle. \quad (11)$$

Then the $\delta_i$ can be interpreted as local distortions as in the adiabatic description, cf. (6). Defining

$$K_{\text{eff}} := \left| \frac{\omega J f}{4g^2}\right|. \quad (12)$$

the equivalence is pushed one step further. Inserting Eq. (12) in Eq. (11) and treating both neighbouring chains as equal yields in CMF the same equation (7) as in the adiabatic treatment. This is true in the D phase and in the I phase as long as the modulation stays close to $\pi$.

The “good” equivalence between the adiabatic treatment and the antiadiabatic treatment with chain-mean-field provides an explanation for the success of the former in spite of the high phonon energies. But it is not the adiabatic treatment in its own right which works so well. There are two major differences between the adiabatic treatment and the seemingly equivalent antiadiabatic treatment. (i) The in-chain couplings are partly phonon-induced and hence they depend on $T$
(ii) In the adiabatic treatment $K$ is related to \textit{in-chain} properties: $K = \omega J/(2g^2)$ (obtained inserting Eq. (5) in $H_{SB}$ and $H_{B}$). The constant $K_{\text{eff}}$ in Eq. (12) refers to an \textit{inter-chain} property since the spatial extent of the phonons as measured by $f$ enters decisively.

What are the points in favour of the antiadiabatic approach for CuGeO$_3$? Besides the relation between $\Delta \approx 25K$ and $\omega \approx 150K$ [11] and the $T$-dependent couplings [28] there is the observation that the zero-point motion of lattice exceeds the static distortions considerably [10]. In particular in the I phase, this zero-point motion has an important effect on the alternating local magnetisation [29,3]. On the basis of results as in Fig. 1 NMR peaks can be understood [30]. In adiabatic calculations the amplitudes of the alternating component, however, is found to be much larger (factor 4 to 6) than in experiment. Theory and experiment can be reconciled if the zero-point motion of lattice exceeds the modulation along the chains. Such translations require that the distortions are also dynamic. So the phasonic zero-point motion provides strong evidence for the nonadiabatic character of the ordered phases in CuGeO$_3$.

2. Incommensurate phase

Turning to the solitons in the I phase we first state the standard theory [31]. The original spin Hamiltonian (6) is mapped by a Jordan-Wigner transformation [32] to a fermionic model which in turn can be reduced to an effective bosonic continuum model at low energies [31,33]

\[ H = \frac{v_S}{2\pi} \int \left( K(\pi \Pi)^2 + \frac{1}{K}(\partial_x \phi)^2 \right) dx \]

\[ + \int \left( -\delta(x) \cos(2\phi) + \frac{K}{2} \delta^2(x) \right) dx \]

where $\delta(x)$ is the alternating component of the distortion. Nakano and Fukuyama used the self-consistent harmonic approximation $\phi \rightarrow \phi_{cl} + \phi_{fl}$ distinguishing a classical number $\phi_{cl}$ and a fluctuating operator part $\phi_{fl}$ with $\langle \phi_{fl} \rangle = 0$. Treating the fluctuation in harmonic approximation leads to $\cos(2\phi) \rightarrow e^{-2\sigma} \cos(2\phi_{cl})(1 - 2\phi_{fl}^2)$ with $\sigma := \langle \phi_{fl}^2(x) \rangle$ motivated by the WKB approach by Dashen \textit{et al.} [34]. Varying the ground state expectation value of (13) with respect to $\delta(x)$ and to $\phi_{cl}$, respectively, yields

\[ 0 = -\cos(2\phi_{cl})e^{-2\sigma} + K\delta \]

\[ 0 = v_S/(\pi K)\partial_x^2 \phi + 2\delta \sin(2\phi_{cl})e^{-2\sigma} \]

which leads finally to

\[ 2\partial_x^2 \phi_{cl} = \sin(2\phi_{cl}) \cos(2\phi_{cl}) \]

where $y = x/\xi$ for the correlation length $\xi$. The \textit{spatial} dependence of $\sigma$ is neglected [31]. Eq. (16) has the soliton array solutions [35,3] (sn, cn, dn: elliptic Jacobi functions)

\[ m_i = W\{dn(r_i/(k_m\xi_m)), k_m)/R \]

\[ +(-1)^i\text{cn}(r_i/(k_m\xi_m), k_m)/2 \]

\[ \delta_i = (-1)^i \delta \text{sn}(r_i/(k_d\xi_d), k_d) \]

with $\xi_m = \xi_d$ and $k_m = k_d$.

In the fits in Fig. 1 one observes that $\xi_m = \xi_d$ does not hold. In addition, there is no good reason to neglect the spatial dependence of $\sigma$ (cf. Ref. [36]). Considering it leads to an extra factor $e^{-4\Delta \sigma}$ on the right side of (16) where $\Delta \sigma$ is the difference to the fluctuations in the ground state. Previous results [4], (not yet self-consistent: fluctuations on top of the Nakano/Fukuyama solution), showed that the spatial dependence of $\Delta \sigma$ is indeed important. The conjecture that the numerical finding $\xi_m = \xi_d$ [3] is related to the spatial dependence of $\Delta \sigma$ could be corroborated.

Figs. 2 and 3 show the results of a fully self-consistent calculation. Clearly the spatial dependence of the fluctuations makes the kink smoother.
This was expected since the fluctuations are maximum in the center of the soliton (see Fig. 4 in Ref. [4]). Most interesting is the fact that the distortion is smoothed more than the alternating magnetisation. From the fits one finds the ratio $\xi_d/\xi_m = 1.24$ in very good agreement with the numerical result at critical frustration $\alpha_c$ (see Fig. 5 in Ref. [3]). Hence the discrepancy between the existing continuum theory of spin-Peierls solitons and the numerical results is removed when the spatial dependence of the fluctuations are considered properly.

In spite of the progress just shown the agreement to experiment is not yet perfect. The soliton widths observed experimentally still exceed the theoretical ones [3]. As a next step towards a quantitative description we include magnetic interchain couplings as they are present and non-negligible in CuGeO$_3$ [37]. They are treated in mean-field approximation so that only the local magnetisations matter. Apart from this extension the Hamiltonian (6) is minimised since the antidiabatic limit leads to the same equations. In Fig. 4 the dependence of the soliton widths on an antiferromagnetic coupling $J_{\text{inter}}$ between adjacent chains in a plane is shown. Clearly, all widths are enhanced. The additional coupling strongly favours the alternating magnetisation close to the center of the soliton. Thus the 2D coupling induces an increase of this region thereby enlarging the soliton [35]. So the agreement with experiment will be improved by taking the 2D character of the magnetic couplings into account. Yet further questions still remain as the ratio between distortive and magnetic soliton width is decreased on increasing $J_{\text{inter}}$ in contrast to experiment.

### 3. Summary

In this work we contrasted the adiabatic approach to spin-Peierls transitions with the antidiabatic approach. If the latter is complemented by a chain-mean-field treatment both approaches yield very similar equations. But the interpretation...
of the parameters differ and so do certain physical phenomena such as $T$-dependent couplings and local magnetisations. For CuGeO$_3$ the antiadiabatic approach was advocated.

In the incommensurate phase the form and the amplitudes of solitons were discussed. The extension of the existing continuum theories as required by the numeric results was presented. Further steps towards a quantitative description of the I phase of CuGeO$_3$ were shown.

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References

[1] J. W. Bray, L. V. Interrante, I. C. Jacobs, and J. C. Bonner, in Extended Linear Chain Compounds, edited by J. S. Miller (Plenum Press, New York, 1983), Vol. 3, p. 353.
[2] J. P. Boucher and L. P. Regnault, J. Phys. I France 6, 1939 (1996).
[3] G. S. Uhrig, F. Schönfeld, J. Boucher, and M. Horvatić, Phys. Rev. B in press (1999).
[4] G. S. Uhrig, in Advances in Solid State Physics, edited by B. Kramer (Vieweg Verlag, Braunschweig, 1999, Vol. 39, p. 291, cond-mat/9904387.
[5] J. Riera and A. Dobry, Phys. Rev. B 51, 16098 (1995).
[6] G. Castilla, S. Chakravarty, and V. J. Emery, Phys. Rev. Lett. 75, 1823 (1995).
[7] K. Fabricius et al., Phys. Rev. B 57, 1102 (1998).
[8] M. Braden et al., Phys. Rev. B 54, 1105 (1996).
[9] M. Braden et al., Phys. Rev. Lett. 80, 3634 (1998).
[10] R. Werner, C. Gros, and M. Braden, Phys. Rev. B 59, 14556 (1999).
[11] G. S. Uhrig, Phys. Rev. B 57, R14004 (1998).
[12] A. Klümpner, R. Raupach, and F. Schönfeld, Phys. Rev. B 59, 3612 (1999).
[13] M. C. Cross and D. S. Fisher, Phys. Rev. B 19, 402 (1979).
[14] L. G. Caron and S. Moukouri, Phys. Rev. Lett. 76, 4050 (1996).
[15] C. Gros and R. Werner, Phys. Rev. B 58, R14677 (1998).
[16] E. Pytte, Phys. Rev. B 10, 2039 (1974); U. Brandt and H. Leschke, Z. Phys. 271, 295 (1974); K. Kuboki and H. Fukuyama, J. Phys. Soc. Jpn. 56, 3126 (1987); F. H. L. Essler, A. M. Tsvelik, and G. Delfino, Phys. Rev. B 56, 11001 (1998).
[17] A. Weiße, G. Wellein, and H. Fehske, cond-mat/9901262.
[18] S. Eggert, Phys. Rev. B 54, R9612 (1996).
[19] R. J. Bursill, R. H. McKenzie, and C. J. Hamer, Phys. Rev. Lett. in press (1999).
[20] D. Augier, D. Poilblanc, E. Sørensen, and I. Affleck, Phys. Rev. B 58, 9110 (1998).
[21] A. W. Sandvik and D. K. Campbell, cond-mat/9902228.
[22] R. W. Kühne and U. Lüöw, cond-mat/9905337.
[23] A. E. Feiguin, J. A. Riera, A. Dobry, and H. A. Ceccatto, Phys. Rev. B 56, 14607 (1997).
[24] F. Schönfeld, G. Bouzerar, G. S. Uhrig, and E. Müller-Hartmann, Eur. Phys. J. B 5, 521 (1998).
[25] P. Hansen, D. Augier, J. Riera, and D. Poilblanc, Phys. Rev. B cond-mat/9805325.
[26] D. Augier et al., cond-mat/9807265.
[27] C. Knetter and G. S. Uhrig, Eur. Phys. J. B in press (1999).
[28] K. Fabricius and U. Lüöw, Phys. Rev. B 57, 13371 (1998).
[29] G. S. Uhrig, F. Schönfeld, and J. Boucher, Europhys. Lett. 41, 431 (1998).
[30] Y. Fagot-Revurat et al., Phys. Rev. Lett. 77, 1861 (1996).
[31] T. Nakano and H. Fukuyama, J. Phys. Soc. Jpn. 49, 1679 (1980); ibid. 50, 2489 (1981).
[32] P. Jordan and E. Wigner, Z. Phys. 47, 42 (1928).
[33] J. von Delft and H. Schoeller, Ann. Physik 7, 225 (1998).
[34] R. F. Dashen, B. Hasslacher, and A. Neveu, Phys. Rev. D 10, 4114 (1974); ibid. 4130; ibid. 4138; ibid. 11 3424 (1975); ibid. 12 2443
[35] J. Zang, S. Chakravarty, and A. R. Bishop, Phys. Rev. B 55, R14705 (1997).
[36] M. Fabrizio, R. Mélin, and J. Souletie, cond-mat/9807094.
[37] G. S. Uhrig, Phys. Rev. Lett. 79, 163 (1997).