Soliton Confinement and the Excitation Spectrum of Spin-Peierls Antiferromagnets

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The excitation spectrum of spin-Peierls antiferromagnets is discussed taking into account phonon dynamics but treating inter-chain elastic couplings in mean field theory. This gives a ladder of soliton-anti-soliton boundstates, with no soliton continuum, until soliton deconfinement takes place at a transition into a non-dimerized phase.

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Much of the theoretical work on spin-Peierls systems largely ignores phonon dynamics, regarding the lattice distortion as being static, thus producing an alternating Heisenberg exchange coupling. In the approach of Cross and Fisher [1] the resulting susceptibility of the spin system (its response to a lattice distortion) is then fed into an RPA calculation of the phonon Green’s function. However, the magnetic excitations, at least in the dimerized phase, are regarded as containing no phonon component. On the other hand, some theoretical work on the magnetic field or impurity induced undimerized phase has considered solitons which are at the same time magnetic excitations (they have spin 1/2) and involve lattice dynamics (the lattice distortion switches between the two phases at the location of the soliton). Khomskii et al. [2] have developed a simple appealing picture of the spin-Peierls transition (as a function of field, impurity concentration or temperature) based on soliton unbinding. Their approach differs fundamentally from that of Cross and Fisher in its treatment of phonon dynamics. Khomskii et al. attempt to treat the one-dimensional phonon dynamics accurately, while approximating the inter-chain elastic couplings by mean field theory. This approach probably works best when the transverse phonon dispersion energy is small compared to the magnetic energy gap. Such an approach leads to an effective one-dimensional model containing both spontaneous dimerization due to one-dimensional phonons and explicit dimerization produced by the mean field of the neighbouring chains. This is to be contrasted with the other approach which only contains explicit dimerization. In the approach of Khomskii et al. the fundamental excitations of the system are solitons. In the spin-Peierls ordered phase the solitons (\(s\)) and anti-solitons (\(\bar{s}\)) are bound together in pairs by a linear potential arising from the explicit dimerization potential of the neighbouring chains. A sufficient temperature, field or impurity concentration drives this self-consistently determined dimerizing field to zero, eliminating the linear potential between solitons, allowing free solitons to propagate. From this perspective the spin-Peierls transition corresponds essentially to soliton deconfinement rather than phonon softening.

In this paper we wish to extend this approach to a more quantum mechanical treatment of the excitations in the spin-Peierls ordered phase. The \(ss\) system, with its linear potential is quantized, leading to a ladder of boundstates which can have spin 0 or 1. The \(s=1\) boundstates correspond to magnons. As the system becomes more one-dimensional, the linear potential gets weaker and the number of stable boundstates increases, diverging in the one-dimensional limit. The \(s=0\) boundstates could also be interpreted as boundstates of two magnons, but in the highly one-dimensional case they are better interpreted as weakly bound singlet \(ss\) pairs with energies given by twice the energy of the soliton plus a (positive) “binding energy” associated with the linear potential. All boundstates are both magnetic and elastic in character; the \(s=0\) boundstates are not necessarily distinct from optical phonons.

Within this approach, a soliton continuum of excitations does not occur in the ordered phase, contrary to the claims in [2]. Instead the continuum is quantized into a ladder of boundstates. A magnon continuum can occur, beginning at precisely twice the magnon gap. All excited \(ss\) boundstates must lie below this continuum (in the appropriate spin channel) in order to be stable. The occurrence of \(ss\) boundstates in Heisenberg models with competing spontaneous and explicit dimerization was mentioned by Haldane [4]. The fact that a soliton continuum cannot occur in spin-Peierls systems due to soliton confinement was pointed out recently by Uhrig and Schulz [5].

The occurrence of a ladder of \(ss\) boundstates is generic to quasi-one-dimensional systems with broken discrete symmetries, at least within this type of mean field treatment. A very similar approach was taken by Shiba [6] to Ising antiferromagnets who referred to the excitations as a “Zeeman ladder”. These excitations have apparently been observed in CsCoCl\(_3\) [7]. Such ladders of boundstates also occur in confining (1+1) dimensional quantum field theories.

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such as quantum electrodynamics (Q.E.D.) \cite{8}, the CP$^n$ model \cite{4} and a generalized “two-harmonic” version of the sine-Gordon model discussed below. Our approach to the spin-Peierls problem was partly inspired by the work of Coleman \cite{8} on weakly coupled Q.E.D.

We begin by considering a simple one-dimensional $s=1/2$ antiferromagnetic model without phonons:

$$
H = \sum_i [J(1 + \delta(1)') \vec{S}_i \cdot \vec{S}_{i+1} + J_2 \vec{S}_i \cdot \vec{S}_{i+1}].
$$

(1)

We first discuss the case $\delta = 0$. For $J_2 < J_{2c} \approx .24J$ the model is in a non-dimerized gapless phase. On the other hand, for larger $J_2$ the groundstate is spontaneously dimerized and there is a gap \cite{10,11}. Thus, in principle, spontaneous dimerization could occur even without phonons. There is some evidence that the value of $J_2$ in CuGeO$_3$ may be close to $J_{2c}$ \cite{11} but clearly phonons also play an important role in the spin-Peierls transition. We include $J_2$ here for a different reason. In order to capture the essential physics of spontaneous dimerization without explicitly including phonons, it is necessary to choose $J_2 > J_{2c}$.

In the limit where $J_2$ is only slightly larger than the critical value the low energy excitation spectrum can be determined by bosonization \cite{12}. This gives the sine-Gordon model with Hamiltonian density:

$$
\mathcal{H} = v \left[ \frac{\beta^2}{8} \Pi^2 + \frac{2}{\beta^2} \left( \frac{d\phi}{dx} \right)^2 + g \cos 2\phi \right],
$$

(2)

with $8\pi - \beta^2$ and $g \propto (J_2/J_{2c} - 1)$. Since this interaction has renormalization group scaling dimension $\beta^2/4\pi$ it is marginally relevant for $J_2 > J_{2c}$ leading to a soliton gap $\Delta_s \propto \exp[-\text{const}/(J_2 - J_{2c})]$. It follows from the bosonization procedure that $\phi$ is to be interpreted as an angular variable and that the discrete symmetry $\phi \rightarrow \phi + \pi$ corresponds to translation by one site. Classically there are two groundstates at $\phi = \pm \pi/2$, corresponding to spontaneously broken translational symmetry. The $s$ and $\bar{s}$ interpolate between these groundstates and have spin $S^z = \pm 1/2$ depending on whether $\phi$ rotates clockwise or counterclockwise with increasing $x$. These groundstates may be simplistically pictured as consisting of nearest neighbour dimers in one of the two possible patterns and the $s$ or $\bar{s}$ as being a single unpaired spin separating the two different dimer patterns, as shown in Figure 1. The (presumably) exact results on the sine-Gordon model indicate that there are no other excitations besides the solitons and anti-solitons (and of course their multi-particle states) for this range of $\beta$ \cite{13,14}. In particular, unlike in a perturbative treatment in $\beta$, there are no approximately harmonic excitations in addition to the topological ones. This is presumably a rather special feature of this particular model. The soliton width and spin correlation length obey $\xi_s \propto v/\Delta_s$, diverging at $J_{2c}$. As we increase $J_2$ further this soliton width decreases. At the special point $J_2 = J/2$ the exact groundstate is given by nearest neighbour dimers. In this entire region the translational symmetry is spontaneously broken so it follows from general principles that the excitation spectrum must contain solitons.

![FIG. 1. An $s\bar{s}$ pair. The solid line represents a dimer singlet.](image)

Next we consider the model of Eq. (1) with a small non-zero $\delta$ corresponding to a mean field from the neighbouring chains preferring one of the two dimerization patterns. The staggered term leads to an additional sine-Gordon interaction:

$$
\mathcal{H} = v \left[ \frac{\beta^2}{8} \Pi^2 + \frac{2}{\beta^2} \left( \frac{d\phi}{dx} \right)^2 + g \cos 2\phi + c\delta \sin \phi \right],
$$

(3)

where $c$ is a constant of $O(1)$. The spectrum is fundamentally different depending on the value of $J_2$. For $J_2 < J_{2c}$ we may ignore the marginally irrelevant $\cos 2\phi$ interaction in Eq. (3) arising from the uniform terms in Eq. (1). On the other hand, the $\sin \phi$ interaction has dimension 1/2 and thus produces a gap $\propto \delta^{2/3}$ (up to logarithmic corrections). The exact spectrum consists of a triplet and a higher energy singlet with $\Delta_1/\Delta_3 = \sqrt{3} \left[3 \right]$. For $J_2 > J_{2c}$, there is a competition between spontaneous and explicit dimerization, represented by the $g \cos 2\phi$ and $c\delta \sin \phi$ terms respectively in the sine-Gordon Hamiltonian. If $\delta$ is very small then, while the true groundstate has $\phi = -\pi/2$ the other state at $\phi = \pi/2$ has only slightly higher energy. Consider a $s\bar{s}$ configuration where the true groundstate ($-\pi/2$) occurs at $x \rightarrow \pm \infty$ but the unstable groundstate occurs in between the $s\bar{s}$ pair, separated
by a distance $x$. Defining the $s$ as the kink with $\phi = -\pi/2$ at $x \to -\infty$ and the $\bar{s}$ as the kink with $\phi = \pi/2$ at $x \to -\infty$, we see that the $s$ must always be to the left of the $\bar{s}$. If $x$ is much greater than the soliton width $\xi$, then the classical energy of such a configuration is simply $2\Delta_s + c\delta x$. This is true regardless of the spins of the two solitons (i.e. whether $\phi$ winds clockwise or counter-clockwise). This linearly confining potential is crucial to the physics of spin-Peierls systems or indeed of any system with a competition between spontaneous and explicit breaking of a discrete symmetry.

We now wish to treat this model quantum mechanically, in the small $\delta$ limit. As far as we know, this “2-harmonic sine-Gordon model” model is not integrable. However, for small $\delta$ we may follow Coleman’s treatment of weakly coupled (1+1)-dimensional Q.E.D. Consider a single $ss$ pair. We expect it to give a spectrum of boundstates, due to the linear potential. Thus we write an effective Hamiltonian for the center of mass co-ordinate, $x$, of the pair:

$$H = -\frac{1}{M_s} \frac{d^2}{dx^2} + c\delta x,$$

where $M_s \approx \Delta_s/v^2$ is the soliton mass. We restrict $x \geq 0$ and impose a vanishing boundary condition on the wavefunction at $x = 0$. The most important point about the eigenstates of this Hamiltonian is that they consist entirely of boundstates; there is no continuum. This follows because the potential keeps on increasing for all $x$ so the soliton and anti-soliton never escape to infinity. A good idea of the nature of the spectrum can be obtained from the WKB approximation. The $n^{th}$ eigenvalue is given by:

$$\int_0^{x_0} dx \sqrt{M_s(E_n^B - c\delta x)} = n\pi,$$

where $x_0$ is the classical turning point, $x_0 \equiv E_n^B/c\delta$. Thus the number of states with energy less than $E^B$ is:

$$N(E^B) \approx \frac{2\sqrt{M_s}(E^B)^{3/2}}{c\delta 3\pi}.$$

We see that the density of states per unit energy diverges as $\delta \to 0$. This is another peculiarity of a linear potential. As the strength of the potential goes to 0 the free particle limit is obtained by the boundstates becoming more and more dense until they fill in the $ss$ continuum. We have attached the superscript on $E^B$ to remind the reader that this is the binding energy of the $ss$ pair; the total energy consists of this binding energy together with the $ss$ rest mass energy:

$$E_n = 2\Delta_s + E_n^B.$$

Actually, the results of the previous paragraph are only valid for low energy boundstates with $E^B << \Delta_s$ where the non-relativistic approximation to the soliton dispersion relation may be used. Coleman extended the validity of this result by using the relativistic version of the WKB approximation. This is certainly valid for Q.E.D. We also expect it to be valid for our antiferromagnetic chain in the limit where $c\delta << \Delta_s << J$, in which the theory is approximately Lorentz invariant up to the energy scale $\Delta_s$. Thus the free soliton energy may be written:

$$E(p) = \sqrt{\Delta_s^2 + v^2p^2}.$$

The WKB condition now becomes:

$$\int dp dx \theta(E_n - 2E(p) - c\delta x) = \frac{1}{c\delta} \int dp |E_n - 2E(p)| \theta[E_n - 2E(p)| = 2\pi n.$$

Here $E_n$ is the full relativistic energy of the $ss$ pair including both kinetic energy of the individual solitons and binding energy. So far this discussion ignores many body effects. In the weak coupling limit, as Coleman observed, these simply truncate this boundstate spectrum at $E < 4\Delta_s$. Any $ss$ boundstate of higher energy is unstable because it can decay into a pair of boundstates. If we imagine trying to pull an $ss$ pair apart to $\infty$ it eventually becomes energetically favourable for “pair production” to occur so that we end up separating to $\infty$ two $ss$ pairs. Setting $E = 4\Delta_s$ in Eq. (9) gives the number of stable boundstates:

$$N \approx \frac{.684\Delta_s}{c\delta}.$$

We note that this $1/\delta$ behavior is independent of the free soliton dispersion relation (although the prefactor depends on it). Shiba encountered essentially a discrete lattice version of this Schroedinger equation in his work on Ising antiferromagnets, coming to similar conclusions about the spectrum.
So far we have ignored the soliton spin. The Hamiltonian of Eq. (4) is spin-independent so both the $s$ and $\bar{s}$ can independently have spin up or down. Thus each of these boundstates corresponds to a degenerate triplet and singlet. When the $s$ and $\bar{s}$ are close together (on the scale $\xi$) their interaction will be a good deal more complicated; in particular it will be spin dependent. Fortunately, for very small $\delta$ they stay far apart even in the lowest boundstate. Note that the $n = 1$ classical turning point is at $x_0 \propto 1/\delta^{1/3}$. As we increase $\delta$ we expect the number of boundstates to decrease and the degeneracy between triplet and singlet boundstates to be lifted. The set of stable boundstates must always lie below the two boundstate continuum in the corresponding spin channel. Clearly free solitons can never appear in the spectrum for non-zero $\delta$. Eventually, when $\delta >> (\Delta_s/J)^{3/2}$ (but still $\delta << 1$) we expect to recover the spectrum of the $J_2 < J_{2c}$ model with one triplet and one singlet with the ratio $\Delta_1/\Delta_3 = \sqrt{3}$. It would be interesting to study the spectrum of the spin model numerically to test these ideas. (The expected behavior for $J_2 < J_{2c}$ and $\delta << 1$ was recently confirmed [14].)

We now include phonons in our model, taking the Hamiltonian:

$$H = \sum_i ((J + \alpha u_i)\vec{S}_i \cdot \vec{S}_{i+1} + \Pi_i^2/M + (K/2)u_i^2 + \delta(-1)^i u_i).$$

Here $u_i$ is the change in separation of two neighbouring ions from its uniform value and $\Pi_i$ is the corresponding conjugate momentum; $M$ is the ionic mass. (We have omitted acoustic phonons for simplicity. They presumably are not important for the spin-Peierls effect.) $\delta$ represents a mean field arising from the coupling to neighbouring chains which favors one of the two possible lattice distortions. We may also keep a second nearest neighbour exchange coupling; it doesn’t change the discussion qualitatively. While this Hamiltonian is considerably more complicated than the spin-only one considered above, we expect many features of the previous discussion to carry over. When $\delta = 0$ we expect two degenerate groundstates with $<u_i> = \pm u_0 (-1)^i$. The excitation spectrum will include s=1/2 solitons and anti-solitons. Note that all excitations now involve both spin and phonon degrees of freedom. In addition to solitons, the excitation spectrum will presumably include other excitations corresponding to optical phonons. In principle there might also be integer spin magnetic quasi-particles below the two-soliton continuum. However, since the interaction between $s$ and $\bar{s}$ vanishes at long distances for $\delta = 0$, no boundstates need occur in this limit, as in the previous spin-only model. Turning on a small $\delta$ will again confine the solitons into $s\bar{s}$ pairs. In the small $\delta$ limit we expect the number of stable boundstates to be proportional to $1/\delta$. Presumably spin 0 $s\bar{s}$ boundstates can mix with optical phonons, which may reduce the number of stable $s=0$ boundstates. As $\delta$ increases the number of boundstates will decrease. Free solitons can never appear in the spectrum. In addition to the stable boundstates there will also be two boundstate continua in the various spin channels (and also presumably a two phonon continuum). Evidently all boundstates must lie below the continuum in the corresponding spin channel in order to be stable. Numerical results on such a one-dimensional spin-phonon system would be highly desirable although considerably more difficult than for a spin-only system.

Finally, let us consider a full three dimensional spin-Peierls Hamiltonian. A simple model would consist of chains with the Hamiltonian of Eq. (1) (with $\delta = 0$) together with an inter-chain phonon coupling:

$$K'\sum_i \sum_{\vec{R},\vec{R'}} u_i\bar{u}_{i\vec{R}}u_{i\bar{R}'}.$$  \hspace{1cm} (12)

where $\vec{R}$ labels the lattice points perpendicular to the chains. In the ordered phase $<u_{R}> = (-1)^i u_0$. Treating the interchain coupling in mean field theory gives the one dimensional model of Eq. (1) with $\delta = ZK'u_0$ where $Z$ is the chain co-ordination number. $u_0$ should then be determined self-consistently by solving the one-dimensional model. This is essentially the approach advocated by Khomskii et al. [3]. Note that it is a rather standard approach to various quasi-one-dimensional systems [15]. In the disordered phase, $\delta = 0$ so solitons can exist as independent excitations. In the ordered phase $\delta > 0$ and free solitons cannot occur due to the confining potential. While based on a mean field treatment of inter-chain couplings this conclusion is presumably much more general. It essentially follows from Landau’s argument that any non-zero density of free solitons leads to the destruction of long range order in a one dimensional system. The theoretical discussion in Ref. [3] essentially considered a purely one-dimensional spin-phonon model which does indeed contain free solitons. The magnon was regarded as an $s\bar{s}$ boundstate that could occur in this model. However, it is not permissible to ignore inter-chain elastic coupling in the dimerized phase. This coupling at the same time stabilizes the dimerized phase up to a finite critical temperature and confines the solitons. Note that in the small $K'$ limit, the energy scale of all excitations is set by the one-dimensional model and therefore the magnon gap should be given by twice the soliton gap (assuming no boundstates in the one-dimensional model for $\delta = 0$). Even if $K'$ is not very small, its effects become smaller near a transition into a non-dimerized phase, driven
by temperature, field or impurity concentration. Thus, at least naively, one might expect the present approach to become more valid near such a transition.

What does this approach tell us about CuGeO$_3$? We may interpret the observed magnon as a spin 1 $s\bar{s}$ boundstate. Likewise, if there is a stable singlet quasi-particle, as suggested by Raman scattering [16], we may interpret it as an $s=0$ $s\bar{s}$ boundstate. As remarked above, there appears to be no sharp distinction, in general, between such an $s=0$ $s\bar{s}$ boundstate and an optical phonon in a spin Peierls system. In Raman scattering the photon couples both to lattice displacements and to $S_i \cdot \bar{S}_{i+1}$. Such spin 0 excitations might also be observable in neutron scattering because they couple to lattice displacements which are excited by neutron scattering from the ionic nuclei. One precise conclusion from the present approach is that there can be no soliton continuum in the spin-Peierls phase. The continuum in the magnetic neutron scattering cross-section necessarily starts at twice the gap to the lowest spin triplet $s\bar{s}$ boundstate corresponding to a 2-magnon continuum. This is consistent with the data presented in [3]. The apparent absence of additional $s\bar{s}$ boundstates in CuGeO$_3$ (besides the magnon and possibly one singlet) can presumably be attributed to the relatively large value of the inter-chain phonon coupling, $K'$. Likewise the failure to observe an $s\bar{s}$ continuum above $T_{SP}$ can be so attributed since then $T_{SP}$ is of the order or greater than $\Delta_s$, smearing the continuum threshold.

It would be interesting to find spin-Peierls materials which were more highly one-dimensional with respect not only to their magnetic exchange couplings but also their elastic couplings. In such materials additional $s\bar{s}$ boundstates should exist below $T_{SP}$ and the $s\bar{s}$ continuum could perhaps be observed above $T_{SP}$. One way in which enhanced one-dimensionality might occur is in a system where the next nearest neighbour Heisenberg coupling $J_2 > J_2c \approx 24J$. In this case the soliton gap could be determined primarily by the magnetic exchange energies and might be large compared to $K'$ or other phonon energy scales.

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