Mixing of magnetic and phononic excitations in incommensurate Spin-Peierls systems

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We analyze the excitation spectra of a spin-phonon coupled chain in the presence of a soliton. This is taken as a microscopic model of a Spin-Peierls material placed in a high magnetic field. We show, by using a semiclassical approximation in the bosonized representation of the spins that a trapped magnetic state obtained in the adiabatic approximation is destroyed by dynamical phonons. Low energy states are phonons trapped by the soliton. When the magnetic gap is smaller than the phonon frequencies the only low energy state is a mixed magneto-phonon state with the energy of the gap. We emphasize that our results are relevant for the Raman spectra of the inorganic Spin-Peierls material CuGeO₃.

The discovery in 1993 by Hase et al [1] of the first inorganic Spin-Peierls compound CuGeO₃ has opened the possibility of study the physics of this collective phenomena in a deep way. Several experimental proves have given an exhaustive information about the excitation spectra of this system and its evolution with an applied magnetic field. The effect of non-magnetic impurities has been investigated also.

Theoretical studies have focused on simplified magnetic model. The excitation spectra in the low temperature phase have been analyzed using a dimerized and frustrated Heisenberg chain as a minimal model for this material [2],[3]. The logic underlying these studies are: the competition between magnetic and elastic energies resolves in the low temperature phase in the dimerization of the lattice. Once this process takes place, phononic and magnetic excitation completely decouple and the magnetic excitations are the same as the chemical dimerized system. This point of view is based on an adiabatic approximation supposing that the energy scale of the magnetic process are high enough respect to the phononic ones. As it has been recently emphasized [3], this relation is not fulfilled for CuGeO₃ where the phonons relevant for the dimerization process are about one order of magnitude more energetic than the magnetic gap. The adiabatic approximation is questionable for this system. An antiadiabatic approach has been developed. The frustrated interaction arises, in this context, from the integration of the in-chain phonons and the explicit dimerization from the interchain interaction treated in a mean field approximation [4]. The same frustrated-dimerized Hamiltonian is therefore obtained but with a reinterpretation of the parameters. What is clearly missed in these studies is a general understanding on how spin and phonons mix as elementary excitation and how the spectra of Spin-Peierls systems is built as a result of this mixing. Some recent numerical results have partially addressed this question [5].

In this paper, we analyze the excitation spectra of one-dimensional spin-phonon system by semiclassical techniques on the bosonized representation of the spins subsystem. We focus on the properties of this system in a high magnetic field. In the dimerized phase the system is in a singlet ground state. Coupling with a magnetic field is not effective up to a critical field value where some dimer breaks and their spins are freely from their singlet. The lattice also relaxes in this process forming the so called soliton lattice. This relaxation is a first indication that spin and phonons do not act as independent excitation. It has been experimentally shown by NMR measurements that the lattice becomes incommensurate following a soliton pattern [6]. The magnetic profile of the soliton has been analyzed by NMR measurement [7].

More recently, optical proves have shown the spectral signature of the incommensurate phase. In the uniform dimerized phase a low energy resonance appears at 30 cm⁻¹ which is a smaller energy than two times the magnon gap [8]. This peak has been adjudicated to a bound state of two triplet [9]. When the incommensurate phase appears the spectral weight is transfered from this state to a lower energy peak at the position of the magnon gap [10]. Direct magnon process could not be seen in the optical response due to spin conservation. Soliton assisted one magnon excitation was recently been proposed as the origin of this peak [11] in similarity with the situation in the presence of non-magnetic impurities [12].

This approach is based on the image of the soliton as an isolated spin in the externally dimerized chain so that an adiabatic approximation is supposed. We will show that this state in fact disappears when the dynamics of the phonons is included. We will also show that a trapped
magneto-phonon state with the energy of the gap appears in the antiadabatic regime so explaining the optical data.

Let us proceed more formally. Our starting Hamiltonian for an Spin-Peierls compound is:

\[ H = H_{ph} + H_{mg} \]

\[ H_{ph} = \sum_i \frac{P_i^2}{2M} + \frac{K}{2}(u_{i+1} - u_i)^2 \]  

\[ H_{mg} = \sum_i J(1 + \frac{\alpha}{2}(u_{i+1} - u_i))\mathbf{S}_i \cdot \mathbf{S}_{i+1} \]

\[ \mathbf{S}_i \] are spin-1/2 operators of the \( i \) ion and \( \alpha \) the magnetoelastic coupling. \( H_{ph} \) represents our simplified model for the phonons. It contains a scalar coordinate \( u_i \) and its conjugate momentum \( P_i \) which are supposed to be the relevant ion coordinates for the dimerization process. As we will only retain the phonons relevant for SP transition, the specific dispersion of the phonons is not important in our approach. The tridimensional character of the phonons field is essential to account for the finite Spin-Peierls temperature and the excitations in the low temperature dimerized phase \[13\]. We will discuss later its effect on the incommensurate phase.

The low energy spectrum could be studied by bosonization. The spin variables are approximately represented by the bosonic field \( \phi(x) \) and its conjugated momentum \( \Pi(x) \) \[14\]. In addition, we retain only the phonon modes producing a smooth deviation of the dimerized pattern. So, we make the replacement \((-1)^i u_i \rightarrow u(x)\). The low energy Hamiltonian becomes:

\[ H_{ph} = \int dx \left\{ \frac{aP(x)^2}{2M} + \frac{2K}{\alpha}u(x)^2 \right\} \]

\[ H_{bos} = \int dx \left\{ \frac{1}{2}v_s^2(\partial_x \phi(x))^2 + \frac{\eta}{2}v_s^2(\partial_x \Pi(x))^2 + \frac{J_0 u(x)}{\pi a_0} \sin \phi(x) \right\} \]

where \( \alpha \) is the lattice constant of the original chain and \( a_0 \) a short range cutoff introduced in the bosonization procedure. \( v_s \) is the spin wave velocity and \( \eta \) is related with the exponent of the correlation functions. For the isotropic Heisenberg model with nn interaction we have \( v_s = \frac{J_0 a_0}{\eta^2} \) and \( \eta = 2 \).

We use a semiclassical approach in the form of a self consistent harmonic approximation(SHA) as it has been originally proposed by Nakano and Fukuyama (NF) \[15\]. In this approach the boson field \( \phi \) is split in a classical component \( \phi_{cl} \) and the quantum fluctuation \( \hat{\phi} \). The last term of \( (4) \) is developed up to second order around \( \phi_{cl} \) and then treated self-consistently by the following replacement:

\[ e^{-\frac{(\hat{\phi})^2}{2}}[\sin(\phi_{cl})(1 - \frac{\hat{\phi}^2}{2}) + \cos(\phi_{cl})\hat{\phi}] \]

\( \langle \hat{\phi}^2 \rangle \) is the ground state expectation value.

In their original work NF have fixed the displacement field \( u \) to its equilibrium classical value \( (u_{cl}) \), so that an adiabatic approximation was assumed. Let us summarize the main results of this study and its consequence for the spectra:

- The classical equations for \( \phi \) and \( u(x) \) have a soliton solution of the form: \( \sin(\phi_{cl}) = \tanh(\lambda x / \xi) \) \( u_{cl}(x) = u_0 \tanh(\lambda x / \xi) \). \( \xi \) is the soliton width, is \( v_s / \Delta \) and \( \Delta \) is the gap over the homogeneous dimerized state (the ‘magnon’ gap) and \( u_0 \) are the displacements in this homogeneous dimerized state. The soliton carries \( S_z = 1 \) spin. In the presence of an external magnetic field these solitons will condense in the ground state and accommodate in a soliton lattice structure.

- The creation energy of the soliton as well as the excitations over this solitonic vacuum could be evaluated once the eigenvalues of the fluctuation operator are known. This eigenvalue problem corresponds to a one of a Schroedinger-like equation which reads as:

\[ \{-\nabla^2 + \tanh^2(\hat{x})\} \psi_\lambda = \Omega(\lambda)^2 \psi_\lambda(\hat{x}) \]

\( \hat{x} = \frac{x}{\xi} \)

\( \Omega(\lambda) \) are the frequency oscillations in presence of the soliton. Eq. \( (6) \) has one bound state at \( \Omega_{cl} = \sqrt{\frac{\sqrt{\frac{v_s}{\Delta}}}{\pi}} \sim 0.786 \) and a continuum started at \( \Omega = \Delta \). The excitation spectra of the theory is spanned by the following state:

- The ground state of the quantum soliton builded around \( \phi_{cl} \). Their creation energy \( (E_s) \) is given by its classical energy plus the difference between the sum of the zero point energies of the oscillators \( (\Omega(\lambda)) \) and the ones in absence of the soliton. This creation energy measures the critical field for the commensurate- incommensurate transition \[13\].

- The excited state of the soliton with energy \( E_s = E_s + \Omega_b \). It is related with the bound state of Eq. \( (6) \).

- Labeling by \( q \) the continuum of level of Eq. \( (4) \), they are \( \Omega(q) = v_s \sqrt{q^2 + 1/\xi^2} \). This is just the kinetic energy of a particle in the soliton free sector. In terms of the original spin chain this is the low energy dispersion of the \( S_z = 0 \) component of a
magnon. Therefore this state corresponds to the scattering of a magnon in presence of the soliton. Moreover, when one of the continuum modes is excited once, we get a two particle magnon-soliton state.

The two last adjudication of states have not been done in the original analysis of the NF work. As our formalism breaks SU(2) symmetry at intermediate stage, the total spin of each state is not directly accessible and should be carefully reanalyzed. We have previously stated that the states below the continuum correspond to the emission of a magnon in the presence of the soliton. Therefore they have total spin-3/2. The bound state has the same total spin as the soliton. Then, it is an optical active mode and it is the analogue in our formalism of the so called soliton assisted magnon process founded in the strong dimerization limit [12].

Moreover, the previous predicted spectra could be tested by numerical exact diagonalization on finite chain. To this end, we solve iteratively the adiabatic equations for \( u_i \) arising from Hamiltonian Eq. 1. The ground state of the spin system was recalculated at each iteration step for \( \frac{1}{L} \rightarrow \infty \). The ratio 0.84 between \( \frac{S_z(S_z+1)}{4} \) and crosses for \( S_z = \frac{3}{2} \). The zero of the energies is chosen at the soliton ground state. Energies are in unit of \( J \).

We discuss in the following two effects that could change our estimated value of the bound state from the SHA method:

1. In going from 1 to 2, we have neglected as NF in their original work a term proportional to \( \cos(2\phi) \) in the bosonized theory. Even though this operator plays an important role in the case of Ising anisotropy, it is a marginally irrelevant operator in the Heisenberg case and therefore it should not change the qualitative features previously discussed. Moreover, NF have studied in a further contribution the effect of this term. Unfortunately, the estimation of the bound state value is strongly dependent on the value of the cutoff in the bosonization procedure \( (\alpha) \) in this situation. If we assume as NF that \( \pi \alpha = a \), the bound state becomes \( \Omega_b \Delta = \sqrt{\frac{3\pi}{2}} \sim 0.93 \). We can conclude as a general fact that the presence of this term tend to increase the bound state value.

2. It has been recently noted that a full self-consistent treatment (where \( \langle \phi^2 \rangle \) in Eq. 8 is not fixed to its value in the ground state) changes the form of the soliton. Moreover, it has been shown in Ref. (20) that this treatment produces a smoother soliton than the one considered here. Therefore, we conclude that this treatment will reduce the bound state value.

In the following we analyze the evolution of the spectra when non-adiabatic effects are included. For simplicity we do not introduce the two additional effects previously discussed. These effects will not affect the prediction of the trapped magneto-phonon state we will find.
**Inclusion of non-adiabatic effects.** To go beyond this static adiabatic approximation in this semi-classical calculation it is necessary to include fluctuations in the displacement field $u(x)$ Therefore, we split $u(x)$ as $u_{cl} + \hat{u}$ and replace it in Eq.\((9)\). The classical equations are the same as before. The fluctuation operator is now a 2 x 2 differential operator with component over $u$ and $\phi$. The eigenvalue problem is now given by:

$$
\Delta^2[-\nabla^2 + \tanh^2(\hat{x})]\psi_1 + \frac{u_0\pi}{\Delta}Kv_s\text{sech}(\hat{x})\psi_2 = \Omega^2\psi_1$$

$$
\omega^2u_0\text{sech}(\hat{x})\psi_1 + \omega^2\psi_2 = \Omega^2\psi_2
$$

(7)

\psi_1\text{ and } \psi_2\text{ are the component of the fluctuation eigenvector over } \phi\text{ and } u\text{ respectively. } \omega = \sqrt{\frac{4\pi K}{\Delta}}\text{ is the phonon frequency at } q = \pi \text{ The eigenfrequency are as previously given by } \Omega. \text{ We obtain from the second equation:}

$$
\psi_2 = \frac{u_0\omega^2\text{sech}(\hat{x})}{\Omega^2 - \omega^2}\psi_1
$$

(8)

Replacing in the first equation we have a kind of Schroedinger equation where the potential depends on the energy. This equation reads:

$$
(-\nabla^2 + \tanh^2(\hat{x}))\psi_1 + \left(\frac{\omega}{\Delta}\right)^2\text{sech}^2(\hat{x})\psi_1 = \left(\frac{\Omega}{\Delta}\right)^2\psi_1
$$

(9)

It is worthy to note that, the only relevant parameter is $\frac{\omega}{\Delta}$. Then, we can follow the evolution of the spectra from the adiabatic to the antiadiabatic limit by moving this parameter.

We have numerically solve Eq.\((9)\). In Fig. \(3\), we show the evolution of the excitation spectra as a function of $\frac{\omega}{\Delta}$. The soliton creation energy has been taken as the zero of the energy.

The main results are:

- There is a zero energy solution which was not present in the adiabatic calculation. This mode arises in the complete translation invariance of the theory. It is nothing but the translational mode of the soliton. In a realistic situation solitons are not isolated, they form a soliton lattice. This zero mode will acquire some dispersion giving rise to the so called phason modes of the soliton lattice. If the magnetic field is not much higher than the critical one the solitons will be rather separate and we can neglect their interference.

- In the direction perpendicular to the magnetic chains, solitons will accommodate in a domain wall, i.e. they form an array of parallel solitons. Their coherency is assured by the interchain elastic coupling. The zero mode as well as all the states of the spectra will have a transversal dispersion, they will depend on the momentum perpendicular to the chains. The spectrum shown in Fig. \(2\) gives the \(q=0\) excitations. We will analyze their dispersive character in a forthcoming work.

- The upper branch corresponds to the excited state of the soliton previously found in the adiabatic approximation. Its energy increases with $\omega$ up to a critical value $\frac{\omega}{\Delta} = \frac{1}{2}$ where this state disappear. For this $\omega$ and $\Omega = \Delta$, the energy dependent potential vanishes. The lattice soliton produces an harmonic potential over the magnetic soliton where it oscillates. In the adiabatic approximation the lattice soliton is frozen and the magnetic one oscillates. For finite $\omega$, the lattice soliton can also move and vibrate with respect to the magnetic deformation. Finally, at the critical value of $\omega$ there is no more a localized vibration and the soliton becomes an unique entity both for the lattice and for the spins. This explains the dispersion of the trapped state at a critical phonon frequency.

- The straight line of slope one in Fig.\(2\) corresponds to $\Omega = \omega$. The eigenstate with this energy corresponds to the excitation of a phonon in the presence of the soliton. The eigenvalues near above are satellite phonons corresponding to phonons trapped by the soliton (i.e. particle vibrations near the soliton). They have only spectral weight on the phonons as it can be seen putting in Eq.\((8)\) $\Omega \sim \omega$. For increasing phonon frequency, most of these eigenvalues lose but the lowest survives acquiring the energy of the magnetic gap. This state is a mixing of a magnetic and a phononic excitation as we show in the following.

- Four Peierls active phonons have been identified for CuGeO$_3$ \([21]\). They have frequencies of 3.12, 6.53,
Their mixed character could be advocated by analyzing separately the magnetic and phononic spectral response. The magnetic Raman operator of a dimerized chain is given by $\mathbf{R}_{mg} = \sum_i (1 + \gamma_{mg}(-1)^i) \mathbf{S}_i \cdot \mathbf{S}_{i+1}$, where $\gamma_{mg}$ is a microscopic parameter. In our bosonized formalism the most relevant contribution comes from the staggered part and it is given by $\frac{\gamma_{mg}}{2\pi a} \sin(\phi)$. By using the SHA given by Eq. (3), retaining the term linear in $\phi$ and developing in the basis of the eigenvalues of the fluctuation operator the magnetic spectral weight $(M_n)$ of an state of energy $\Omega_n$ is given by:

$$M_n = \frac{\gamma^2_{mg}}{2\pi_n} \left\{ \frac{4K}{J \alpha} \frac{\Omega_n - \omega^2}{\omega^2} \int \psi_2 dx \right\}^2 \tag{10}$$

where we have used Eq. (3) to write $\psi_1$ as a function of $\psi_2$. Phonons contributions to Raman scattering is proportional to the square of the transition elements of the normal coordinates $\mathbf{R}_{ph} = \gamma_{ph} \int \frac{dx}{a} u(x)$, the phonons spectral weight of the $n$-state is given by:

$$P_n = \frac{\gamma^2_{ph}}{2\pi_n} \left\{ \int \psi_2 \frac{dx}{a} \right\}^2 \tag{11}$$

The relation between the magnetic and phononic contribution to the $n$-th peak of the spectra is:

$$\frac{M_n}{P_n} = \frac{\gamma^2_{mg}}{\gamma^2_{ph}} \left\{ \frac{4K}{J \alpha} \right\}^2 \left\{ \frac{\Omega_n - \omega^2}{\omega^2} \right\}^2 \tag{12}$$

We take the first two factors as a measure of the spin-phonon coupling, we fix this parameters when moving $\omega$. Eq. (12) shows that the magnetic component of a given Raman peak increases when its position shifts from a phonon frequency. The lower energy eigenvalue of Fig. 2 starts being a trapped phonons for $\frac{\omega}{\Omega} < 1$ and transmutes in a magneto-phonon in the opposite limit where the real material lives. Its existence is a consequence of the quasi-one-dimensional character of our system because a one-dimensional well has always one bound state.

In summary, we have shown that nonadiabatic effects are relevant for incommensurate Spin-Peierls system. In the antiadiabatic limit applicable to CuGeO$_3$ we show the apparition of a trapped mixed state with the energy of the magnon gap. It gives an alternative explanation of the Raman induced peak found in the incommensurate phase of CuGeO$_3$. A.D. acknowledges G. Uhrig and A. Greco for useful discussion. We are grateful to J. Riera for useful discussions and computational help. We are grateful to P. Lemmens for discussions and for pointing out Ref. 3.

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