Incommensurate spin Luttinger liquid phase in a model for the spin-Peierls materials TiOBr and TiOCl

D. Mastrogiuseppe, C. Gazza, and A. Dobry
Facultad de Ciencias Exactas Ingeniería y Agrimensura, Universidad Nacional de Rosario and Instituto de Física Rosario, Bv. 27 de Febrero 210 bis, 2000 Rosario, Argentina.
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In the present work we aim to characterize the lattice configurations and the magnetic behavior in the incommensurate phase of spin-Peierls systems. This phase emerges when the magnetic exchange interaction is coupled to the distortions of an underlying triangular lattice and has its experimental realization in the quasi-one-dimensional compound family TiOX (X = Cl, Br). With a simple model of spin-1/2 chains inserted in a planar triangular geometry which couples them elastically, we are able to obtain the uniform-incommensurate and incommensurate-dimerized phase transitions seen in these compounds. Moreover, we follow the evolution of the wave-vector of the distortions with temperature inside the incommensurate phase. Finally, we predict gapless spin excitations for the intermediate phase of TiOX compounds along with incommensurate spin-spin correlations, offering to experimentalists a candidate to observe an exotic Luttinger liquid-like behavior.

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I. INTRODUCTION

The recent discovery of a new class of inorganic spin-Peierls (SP) materials, TiOX (X = Cl, Br), has made reemerge the interest on this magneto-elastic transition. Unlike the previously known SP materials, these compounds present a novel intermediate temperature incommensurate phase between the usual uniform and dimerized ones. Other peculiar properties are the presence of a pseudo spin gap up to $T^* \approx 135 K$ for TiOCl and the existence of a very large singlet-triplet energy gap in the dimerized phase with values $E_g \approx 430 K$ for TiOCl and $250K$ for TiOBr.

The magnetic Ti chains are arranged in a triangular lattice forming a bi-layer structure. Good Bonner-Fisher fittings to magnetic susceptibility measurements are obtained from which it is inferred that these compounds are well described by one-dimensional $S = 1/2$ Heisenberg chains. Moreover, the exchange interaction constants which emerge from these fittings are $J \sim 660 K$ and $\sim 370 K$ for TiOCl and TiOBr respectively. These isostructural compounds present two phase transitions: a uniform-incommensurate one at $T_{c2} \sim 92 K$ and $\sim 48 K$, and an incommensurate-dimerized one at $T_{c1} \sim 67 K$ and $\sim 27 K$ respectively. The order of the high temperature transition has been a subject of extensive controversy although now there appear to be enough evidence that it is a second order transition whereas that at $T_{c1}$ is of first order.

We have recently presented a simple microscopic model of independent Heisenberg chains immersed in a two-dimensional triangular elastic lattice. This model predicts the appearance of the uniform to incommensurate phase transition through the softening of a phonon near the Brillouin zone boundary. This points to the relevant role of the lattice geometry as a driving mechanism for the incommensuration. On the other hand, a precise characterization of the magnetism inside the incommensurate phase is still lacking due to the difficulty of obtaining large enough single crystals to perform inelastic neutron scattering experiments. Recently, the dynamical structure factor of TiOBr was obtained by time-of-flight neutron scattering on powder samples. These measurements suggest the closing of the magnetic gap when going into the incommensurate phase from the dimerized phase. This seems to be in agreement with our theoretical predictions which will be detailed in the present work.

In the first part of this article we do a review of the mechanism leading to the incommensurate transition. The model of Ref. is extended to account for incommensuration in the direction perpendicular to the magnetic chains as well. Within a model of XY spins coupled to adiabatic phonons, we obtain lattice configurations for different temperatures in the incommensurate phase. In the second part, we aim to characterize the magnetic excitations in this phase. We search for the existence or absence of spin gap and calculate spin-spin correlations functions with analytic and numerical techniques.

II. LATTICE GEOMETRY AS A DRIVING FORCE FOR THE INCOMMENSURATION

It is well known that a magnetic chain in which the spins are coupled to the lattice is unstable toward a dimerization of the ions. This is called a spin-Peierls transition. For a real compound to exhibit this kind of ordering, quasi-one dimensionality is needed because large interchain magnetic interactions would make the system develop magnetic ordering before going into the singlet ground state. This is a possible reason why few SP materials are known. Before the discovery of this transition in the TiOX family in 2003, the only known inorganic...
SP compound was CuGeO$_3$. This compound presents a magnetoelastic transition from the uniform phase at high temperatures to a dimerized phase as it is predicted for a single chain. So the question is the following: what makes TiOX behave differently so as to develop an incommensurate phase in between?

Let us consider the spring-like lattice model of Fig. 1 as representative of the Ti ions in the bilayer structure of TiOX.

For simplicity, suppose that the ions move only in the direction of the magnetic chains. Fig. 2 clearly shows a marked difference between the situation where the ions in neighboring chains are aligned normally and that in which they are shifted by half lattice constant.

If the ions dimerize in an in-phase pattern in the square lattice as shown in Fig. 2a, there is no cost of elastic energy because the distance to the corresponding ion in the next chain remains unchanged. Therefore, the system will prefer this kind of distortion to lower its overall free energy. This is what happens in the case of CuGeO$_3$. On the other hand, for the triangular geometry (Fig. 2b) one sees that there is always an increase of interchain energy due to the alternating contraction and expansion of the interchain ionic distances. Thus, it is less favorable for the triangular system to dimerize as it has to pay more elastic energy than in the square case.

Let us perform a more precise characterization of what would happen with the distortions in the triangular case. Now we are going to allow for movements also in the $x$ direction (perpendicular to the magnetic chains) for more generality. The total mean energy of the elastic system can be calculated as $E_{ph} = \frac{1}{2} \sum_{\mathbf{q}} \Omega^2(\mathbf{q})|Q(\mathbf{q})|^2$ where $\Omega(\mathbf{q})$ is the phonon dispersion relation and $Q(\mathbf{q})$ are the normal coordinates. We want to study what happens with the elastic energy as we move away from a in-phase dimerization, i.e. from the component $\mathbf{q} = (0, \pi)$. The square of the lowest dispersion curve $\Omega$ for our model of Fig. 1 (see the phononic Hamiltonian $H_{ph}$ in Eq. 3) reads

$$
\Omega^2(\mathbf{q}) = \Omega_1^2 (1 - \cos q_y) + 2\Omega_2^2 \left( 1 - \cos \frac{q_x}{2} \cos \frac{q_y}{2} \right) + \Omega_3^2 (1 - \cos q_x) - \sqrt{\Omega_1^2 (1 - \cos q_y) + \Omega_3^2 (1 - \cos q_x)}^2 + 4\Omega_4^2 \sin \frac{q_x}{2} \sin \frac{q_y}{2}
$$

where $\Omega_4^2 \equiv 4K_\alpha/M$. If we develop this relation near $\mathbf{q} = (0, \pi)$ we get

$$
\Omega^2(\mathbf{q}) \sim \Omega_2^2 \left[ \left( \frac{q_y^2}{8} + 1 \right) \delta + \left( 2 + \frac{\Omega_4^2}{\Omega_2^2} - \frac{\Omega_3^2}{4\Omega_2^2} \right) q_x^2 \right],
$$

where $\delta \equiv \pi - q_y$.

Therefore we see that the development has a linear term in $\delta$ (or $q_y$) in contrast to what happens for the square lattice in which the first non vanishing $q_y$ term is of second order. To first order, the square lattice does not alter its elastic energy in the vicinity of $\delta = 0$ but there is an energy gain for the triangular lattice as long as the condition $q_x < 2\sqrt{2}$ is fulfilled. This is not a very restrictive condition as we are looking for solutions with $q_x \sim 0$, i.e. a smooth interchain modulation. As $q_y$ is the component which couples to magnetism in our model, then if the magnetic free energy does not grow very rapidly with $\delta$ we expect the total free energy to have a minimum shifted from $\delta = 0$. On the other hand, the first non vanishing $q_x$ term appears at second order. Anyway, for a fixed $\delta > 0$, the system would gain elastic energy moving form $q_x = 0$ if the condition $\Omega_3^2 < \frac{\Omega_2^2}{8} \left( \frac{2q_x^2}{\Omega_4^2} - \delta \right)$.
is satisfied. In conclusion, this model would enable us to find an incommensuration in both $q$-directions as seen experimentally.

For a quantitative characterization of the uniform-incommensurate transition, let us extend our model of Ref. 6 allowing ion displacements in the $x$ direction as well. We introduce the following Hamiltonian,

$$H = H_{ph} + H_s + H_{sph},$$

$$H_{ph} = \sum_{i,j} \frac{P_{i,j}^2}{2m} +$$

$$\sum_{i,j} \left[ \frac{K_1}{2}(u_{i,j}^x - u_{i+1,j}^x)^2 + \frac{K_2}{2}(u_{i,j}^y - u_{i,j+1}^y)^2 \right] +$$

$$\sum_{\alpha=\{x,y\}} \left[ (u_{i,j}^\alpha - u_{i,j+1}^\alpha)(u_{i,j}^\alpha - u_{i,j+1}^\alpha) - (u_{i,j}^\alpha - u_{i-1,j}^\alpha)(u_{i,j}^\alpha - u_{i+1,j}^\alpha) \right],$$

$$H_s = J \sum_{i,j} S_{i,j} \cdot S_{i+1,j},$$

$$H_{sph} = \sum_{i,j} \alpha(u_{i+1,j}^y - u_{i,j}^y) S_{i,j} \cdot S_{i+1,j},$$

where $P_{i,j}$ is the momentum of the $i$-th ion on the $j$-th chain, $K_1$, $K_2$ and $K_3$ are the elastic constants shown in Fig. 4 and $\alpha$ is the spin-phonon coupling constant. The spin-phonon part has been linearized, so the magnetism couples only to the $y$ component of the ion movements to this order. It is useful to introduce a dimensionless spin-phonon constant through

$$\lambda = \frac{4\alpha^2 J}{\pi M \Omega^2(0, \pi)}.$$  \hfill (7)

The phononic part of this Hamiltonian allowed us to calculate the dispersion relation [1]. Following the steps of Ref. 6, we calculate the phononic retarded Green’s function. The phononic self-energy is treated by a Random phase approximation approach and we use the expression obtained in Ref. 6 for the dimer-dimer correlation function. There will be a structural transition when $D^{\text{ret}}(q, \omega)$ has a pole with $\omega = 0$. This gives us an equation for the transition temperature as a function of $q$. The $q$ which has the highest temperature will be the wave vector associated to the lattice distortion at the transition point. Fig. 5 shows a contour map of the frequencies of the softened phononic dispersion surface at the transition temperature, in which it is seen that the mode which drives the structural transition is incommensurate for both $q$-components, in agreement with the experimental measurements [9–13].

The free parameters $K_1$, $K_2$, $K_3$, $\lambda$ of our model have been adjusted in order to fit different experimental data measured in TiOCl (cf. Fig. 3 of Ref. 6): the phonon frequencies obtained by inelastic x-ray scattering for the $(0, q_y)$ direction at $T = 300K$, the value of the bare frequency of the phonon identified as the one which drives the SP transition $\Omega_0 \sim 27\text{meV}$, and $(q_x, q_y)$ values similar to those found experimentally [9,10,13]. Specifically, the values which gave us the best fit were: $\Omega_1 = \sqrt{4K_1/K_2} \sim 6\text{meV}$, $K_2/K_1 \sim 6.7$, $K_3/K_1 \sim 11$ and $\lambda = 1.3$. It is worth saying that this approach overestimates the transition temperature by a factor $\sim 2.5$ as discussed in Ref. 6. We treated this problem in Ref. 14 showing that an approach using non-adiabatic phonons solves the problem, but we will not do the calculations for the present dispersion relation since it does not lead to qualitatively different results.

Now let us turn to follow the evolution of the distortions for different temperatures inside the incommensurate phase. For this purpose we propose as a model an array of XY spin chains immersed on the elastic structure of Fig. 1. As the XY spin-phonon chain contains the relevant physics of a SP system, we expect that the qualitative behavior will be captured by this simplified model, which incidentally can be solved exactly. The Hamiltonian is the one defined in (3) but now we neglect the $z$-component of the spin operators. Now we aim to minimize the total free energy (magnetic + elastic) self-consistently in the displacements at different temperatures. For this purpose we first perform a Jordan-Wigner transformation on the spin operators of this XY Hamiltonian, which leads to a tight-binding-like model of spinless fermions coupled to the lattice. This model can be solved exactly by diagonalizing $N_c$ matrices of size $N_s \times N_s$ (where $N_s$, $N_c$ are the number of sites and chains) each one, and whose elements depend on the ac-
tual displacement configuration. This allows us to obtain the full one-particle spectrum. Then, the free energy per site is obtained by the expression

\[ f = e_{ph} - \frac{T}{N_s N_c} \sum_{l=1}^{N_s} \sum_{m=1}^{N_c} \ln(1 + e^{-\lambda_l^m / T}) \]  

(8)

where \( e_{ph} \) is the elastic energy density associated with \( H_{ph} \) in Eq. 3 and \( \lambda_l^m \) is the m-th eigenvalue of the matrix associated to the l-th chain. By this minimization procedure we are able to obtain both phase transitions, i.e. we can account for the full intermediate incommensurate phase. Fig. 4 shows two examples of distortions of one of the chains in the incommensurate phase, one near the dimerized phase and the other near the uniform one. Superimposed to each pattern, we show the wave which modulates the alternation \((-1)^i u_i\), together with a sinusoidal fit to it.

![Distortion patterns in the incommensurate phase](image)

FIG. 4. Distortion patterns in the incommensurate phase, a) close to the uniform phase at \( T/J \sim 0.14 \) and b) near the dimerized phase at \( T/J \sim 0.12 \). Solid lines represent the waves that modulate the distortions given by \((-1)^i u_i\). Dashed lines are sinusoidal fits to the envelopes.

We observe that in the case close to the dimerized phase, the modulation is not so well described by a sine function. The distortion pattern looks more like a soliton lattice which is confirmed by the secondary peaks appearing in the Fourier transform of the positions (see Fig. 5). When the temperature raises, the modulation evolves and close to the uniform phase it is a perfect sine function (Fig. 4a). This behavior could explain the experimental temperature evolution of the x-ray peaks observed in Ref. [8] for the incommensurate phase of TiOCl. There, a third harmonic of the main incommensurate peak was observed and interpreted as due to a soliton lattice structure. Upon warming toward the uniform phase this third harmonic signal reduces and the pattern becomes sinusoidal. On the other hand, this behavior is also similar to what it is observed in CuGeO\(_3\) when a magnetic field is applied to the compound in the low temperature phase [15]. In that system, when the magnetic field is high enough to overcome the spin gap (first critical field), the system first develops a soliton lattice and with increasing field it tends to a sinusoidal pattern up to a second critical field, above which it becomes uniform.

To finish this section, in Fig. 6 we show the evolution with temperature of the wave vector of the main Fourier peak.

![Evolution of δ and qx with temperature](image)

FIG. 6. Evolution of \( \delta \) and \( q_x \) with temperature. Dashed lines are the frontiers which separate the different phases.

We were able to obtain a similar dependency of \( q_y \) with \( T \) as seen experimentally[9,10]. On the other hand, the \( q_x \) modulation that we obtain increases slightly with decreasing temperature, contrary to what it is found experimentally. This might be due to the fact that our model does not consider magnetic interchain interactions, so the system is not able to gain magnetic energy by a distortion in the x direction, in contrast to what occurs in the direction of the magnetic chains. Fig. 7 shows the evolution of the amplitude of the distortion \( u_0 \), in which we can observe that above \( T_{c2} \) we have \( u_0 = 0 \) indicating that this is the order parameter for the uniform-incommensurate transition.

The behavior of this curve around \( T_{c2} \) resembles that of a second order phase transition (cf. with Ref. [3]). On
the other hand, at $T_{c1}$ there is a finite jump of $u_0$ and $q$ (Fig. 4), signaling a first order transition. The order parameter for this transition is $\delta$.

### III. NATURE OF THE EXCITATIONS AND SPIN CORRELATIONS

Let us now characterize the behavior of the magnetic excitations inside the incommensurate phase. We are going to check for the existence or not of a spin gap and calculate spin-spin correlation functions. For this purpose we tackle the problem with two different approaches.

In the previous section we have minimized the free energy for a system of elastically coupled XY chains and obtained the corresponding distortions for different temperatures. Now we are going to use these configurations as stable patterns for one chain and study some properties on it. First we will check the energy spectrum around the Fermi energy of the pseudo-fermion chains with hoppings modulated by these distortions. As different $S_z$ subspaces in the magnetic problem are related to a different filling of the one-particle energy levels in the spinless fermions, this will enable us to identify the presence or absence of a spin gap. In Fig. 8 we see the spectra for two different patterns. In both cases continuous bands appear at the Fermi level, indicating gapless excitations. We were able to check this behavior for several different patterns which always led to gapless spin excitations.

Now we turn to the calculation of spin-spin correlations $\langle S_i^x S_{i+r}^x \rangle = \langle S_i^y S_{i+r}^y \rangle$. This is accomplished through the calculation of $r \times r$ determinants whose elements are obtained by the formula

$$G_{i,j} = \sum_{m=1}^{N/2} \left( e_i^m e_j^m - e_i^{m+N/2} e_j^{m+N/2} \right)$$

where $e_i^m$ is the $i$-th component of the eigenvector corresponding to the $m$-th eigenvalue of the matrix introduced in the previous section. Fig. 9 shows real space correlations and Fig. 10 shows the static structure factor along with the Fourier transform of the corresponding distortion pattern.

On one hand we see that the spin correlations in the incommensurate phase are modulated. This is made clear by the appearance of new peaks in the structure factor. In every case there appear peaks at $q_y = \pi - 2\delta$ and $q_y = \delta$ (not shown in Fig. 10) besides the one at $q_y = \pi$ which already belongs to the uniform antiferromagnetic lattice. With this information in hand we were able to fit the real space correlations by the following formula

$$\langle S_i^x S_{i+r}^x \rangle = a_0 \cos \delta r \sqrt{r} + a_1 \frac{(1 + a_2 \cos 2\delta r)}{\sqrt{r}} (10)$$

where the parameters $a_i$, which measure the relative intensity of each peak, take on different values for each distortion. On the other hand, the magnetic Fourier peak near $\pi$ is at twice the distance from $\pi$ of the lattice peak.
It is worth mentioning that the peak at \( q_\theta = \delta \) has a much smaller intensity than those at \( q_\theta = \pi \) or \( q_\theta = \pi - 2\delta \).

We now turn to the study of the Heisenberg model with modulated exchange on a single chain. Nowadays, the Density Matrix Renormalization Group (DMRG) numerical technique appears as the ineluctable numerical tool for treating static properties in one dimensional interacting systems at zero temperature\(^{17-21}\). For our purpose we chose its finite size algorithm with periodic boundary conditions (PBC) on systems of sizes up to \( L = 200 \), instead of open boundary conditions to avoid edge effects. Even though it is known that using PBC leads to a slower convergence of the method\(^{12,13}\), keeping just 300 states was enough to get quasi-exact results with a discarded weight of order \( O(10^{-12}) \). In particular we are interested in the spin gap, here determined as \( \Delta_z = E_0(S_z = 1) - E_0(S_z = 0), \) being \( E_0(S_z) \) the ground state energy on a chain in the subspace of a given total \( S_z \). Different single-chain distortion patterns of the type \( u_i = u_0 \cos(q \cdot R_i) \) were tried and the absence of spin gap was found for every configuration, confirming a spin gapless situation in the incommensurate phase. Spin-spin correlation functions have been also calculated and the results are shown in Figs. 11 and 12.

As for the case of the XY model, the real space correlations show a power law behavior, indicative of the absence of spin gap. The decay is modulated by an envelope whose Fourier component is \( q = \pi - 2\delta \) as found previously. We propose, as a check for this prediction, the measurement of the static magnetic structure factor by elastic neutron scattering. This should present a commensurate peak at twice the distance from the incommensurate peak of the low temperature phase with respect to that seen in the lattice structure factor.

To conclude this section, let us comment on the results given by bosonization on the Heisenberg Hamiltonian and a Renormalization Group analysis. This procedure has been treated in Ref. 19 for a problem of fermions with incommensuration. Here, we reproduce the bosonized version of the Hamiltonian using our parameters, which

\[
H = \frac{1}{2\pi_0} \int_0^{2\pi} dx \left[ u K (\pi \Pi(x))^2 + \frac{u}{K} (\nabla \phi(x))^2 \right] \\
+ \frac{c u_0}{2\pi\bar{\alpha}} \int_0^{2\pi} dx \left[ \sin(2\phi + \delta x) + \sin(2\phi - \delta x) \right]
\]

where \( \phi \) is a smooth bosonic field, \( \Pi \) is the field conjugated to \( \phi \), \( u \) is a velocity, \( K \) is the Luttinger parameter and \( \bar{\alpha} \) is a cutoff of the theory.

The flow equation for \( \delta \) is given by \( \frac{d\delta}{dt} = \delta \) (cf. Eq. 45 of Ref. 20). It is easy to observe that \( \delta \) grows with scale, which means that the sine functions in (11) oscillate rapidly so the integral will vanish. Therefore we end up with a free bosonic theory indicating that the excitations will be gapless as in a uniform Heisenberg chain. Note that the bosonized Hamiltonian of Eq. (11) could correspond to a general XXZ model. The only change is the value of the Luttinger parameter \( K \). Therefore, the results presented in this Section seem to indicate that the behavior of the correlation function given in Eq. (11) has an universal form which is obtained by changing the exponent of the power law decay from a square root in the XX model, to a \( K \)-dependent exponent in a general
IV. CONCLUSIONS

In conclusion, we have studied the behavior of a system of spin-1/2 magnetic chains which are immersed in a triangular planar lattice. It is seen that this triangular geometry leads the system to a quite different behavior compared to a regular spin-Peierls mechanism, like that found in CuGeO$_3$ and other organic compounds. The elastic frustration produced by the triangular arrangement of the magnetic ions leads the system to produce an incommensurate phase between the uniform and dimerized ones. This is what it is observed experimentally in the compound family TiOX (X = Cl, Br). We have seen that with a simple model of magnetically uncoupled chains we were able to obtain both phase transitions. Moreover, by a self-consistent minimization of the free energy, we could describe the lattice configurations at different temperatures which allowed us to follow the $y$-component of the main modulation wave-vector with temperature. On the other hand, spin-spin correlation functions were calculated exactly with XY chains and by DMRG calculations on the Heisenberg model on single chains with sinusoidal modulations. In both approaches we obtained power law behaviors at long distances, modulated by an envelope which has a Fourier peak at $2\delta$, where at $\delta$ is the peak of the imposed lattice modulation. Moreover, this power law decay of the correlations is indicative of the absence of spin gap in the excitation spectrum, which was confirmed by looking at the one-particle spectrum in the XY case, and the vanishing energy difference between $S_z = 0$ and $S_z = 1$ subspaces in the case of the DMRG calculations. Furthermore, an analysis of the bosonized version of the spin-phonon Hamiltonian together with the Renormalization Group flow equation of the incommensuration parameter $\delta$ led to the same conclusion. In recent time-of-flight neutron scattering experiments in TiOBr, it has been shown that the spectral weight increases at low energy when going from the dimerized to the incommensurate phase. This is in agreement with the theoretical prediction of the present paper that the gap closes in the incommensurate phase. A more detailed experimental determination of the magnetic spectrum and the theoretical calculation of the dynamical correlation function in the incommensurate phase would be needed to further clarify the nature of the magnetic excitations in this uncommon phase which we have characterized as an incommensurate spin Luttinger liquid phase.

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