

## Anharmonic effects in magnetoelastic chains

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We describe a new mechanism leading to the formation of rational magnetization plateau phases, which is mainly due to the anharmonic spin-phonon coupling. This anharmonicity produces plateaux in the magnetization curve at unexpected values of the magnetization without explicit magnetic frustration in the Hamiltonian and without an explicit breaking of the translational symmetry. These plateau phases are accompanied by magneto-elastic deformations, which are not present in the harmonic case.

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### I. INTRODUCTION

Coupling of electronic and elastic modes has been shown to play a crucial role in many condensed matter systems, most notably in the BCS theory of superconductivity where the presence of the lattice degrees of freedom is crucial to explain pair formation.<sup>1</sup> Another paradigmatic case is the so-called Peierls effect, where modulations in the charge or spin densities may appear due to the electron-phonon interactions (see, e.g., Ref. 2 and references therein). More recently, phonon effects have been observed in many other strongly correlated systems, in particular in some magnetic systems that show plateaux in their magnetization curves.<sup>3</sup>

Usually one expects to have an accurate description of an electron-phonon system by approximating the phonon potential with a quadratic function of the interatomic distances between nearest-neighbor ions on sites  $i$  and  $j$ ,  $\delta_{ij}$ . Within the same degree of accuracy, the dependence in  $\delta_{ij}$  of the hopping amplitudes and/or the magnetic exchange constants is approximated as a linear function. This description works well in most of the cases, since interatomic displacements are usually rather small as has been verified experimentally in many systems, like in the BCS superconductors. More recently, however, a less conventional BCS superconductor, MgB<sub>2</sub>, has shown an unusually high critical temperature, around 40 K, which could be the consequence of strong anharmonicities both in the phonon potential and in the electron-phonon coupling.<sup>4-7</sup>

The relevance of anharmonic couplings has also been discussed in relation to a great variety of compounds, both from an experimental<sup>8-10</sup> and a theoretical point of view,<sup>11</sup> including the family of pyrochlore oxide superconductors, AOs<sub>2</sub>O<sub>6</sub> for  $A = \text{Cs, Rb, and K}$ ,<sup>8</sup> the heavy fermion superconductors PrOs<sub>4</sub>Sb<sub>12</sub> and SmOs<sub>4</sub>Sb<sub>12</sub>,<sup>9</sup> and some potentially thermoelectric materials such as  $X_8\text{Ga}_{16}\text{Ge}_{30}$  ( $X = \text{Eu, Sr, and Ba}$ ),<sup>10</sup> etc. Another possible relevance of anharmonicities is in the study of spin systems in high pulsed magnetic fields and Raman experiments,<sup>12</sup>

Apart from possible experimental motivations, the role of anharmonicities in the physics of low-dimensional systems is interesting in its own right and we investigate this issue in the present paper in one of the simplest and most paradigmatic one-dimensional systems: the  $XXZ$  Heisenberg chain.

More precisely, we analyze in the present paper the effects of anharmonic (adiabatic) phonons in the spin-Peierls mechanism as well as the consequences on the magnetic properties of the  $XXZ$  Heisenberg chain coupled nonlinearly to lattice deformations. The most important consequence of the anharmonicity is that it produces plateaux in the magnetization curve at unexpected values of the magnetization. For example, a plateau at  $M = 1/3$  of saturation magnetization appears without explicit magnetic frustration in the Hamiltonian and without an explicit breaking of the translational symmetry.<sup>13,14</sup> Besides, magnetoelastic deformations appear in some particular cases with frequencies that halve that of the first harmonic,  $2k_F$ , as, e.g., at  $M = 1/5$  (see below). Similar conclusions should apply to more complicated models, since the effects of other interactions such as, e.g., a next-nearest-neighbor interaction would be simply to enlarge the extension of the plateaux phases and to modify the magnitude of the spin gaps.<sup>15</sup>

### II. MODEL

We start from the following spin-phonon Hamiltonian in the limit of large ionic mass  $M \rightarrow \infty$ , the so-called adiabatic limit

$$\mathcal{H} = J \sum_i (1 + A_1 \delta_i + A_2 \delta_i^2) \vec{S}_i \cdot \vec{S}_{i+1} - h \sum_i S_i^z + \sum_i V(\delta_i). \quad (1)$$

Here  $\delta_i$  denotes the interatomic distance between site  $i$  and  $i + 1$ ,  $h$  is the external magnetic field, and  $\vec{S}_i$  are spin-1/2 operators.

The dependence of the spin-phonon coupling on the interatomic distance  $\delta_i$  has been expanded up to second order with coefficient  $A_2$ . A Zeeman term is included to take into account magnetic-field effects.

The phonon potential energy in Eq. (1) is given by

$$V(\delta_i) = \omega_0 \left( \frac{1}{2} \delta_i^2 + \alpha_3 \delta_i^3 + \alpha_4 \delta_i^4 \right), \quad (2)$$

where  $\alpha_3$  and  $\alpha_4$  take into account the anharmonicity of the interatomic potential energy.

Generally, the properties due to the anharmonic oscillations arise both from the addition of quartic terms in the potential energy and next-to-leading terms in the spin-phonon coupling. In this paper, we focus on the contribution of the anharmonicity in the spin-phonon coupling measured by  $A_2$ , ignoring the contribution of higher-order terms in the potential energy. We show that it is the term quadratic in the lattice deformations in the interaction Hamiltonian that changes drastically the physics of the magnetoelastic  $XXZ$  chain. We expect that higher-order terms in the potential energy (cubic and quartic) are inessential.

### III. BOSONIZATION DESCRIPTION

Following the usual procedure in the low-energy limit, we bosonize the spin degrees of freedom at fixed magnetization  $M$  and the interaction term becomes<sup>15</sup>

$$H_{\text{sp-ph}} = \int dx [A_1 \delta_M(x) + A_2 \delta_M(x)^2] \rho(x), \quad (3)$$

where we have introduced the subscript  $M$  in  $\delta_M(x)$  to stress its dependence on the magnetization. Here  $\rho(x)$  is the continuum expression of the energy density

$$\rho(x) = \alpha \partial_x \phi + \beta \cos(2k_F x + \sqrt{2\pi} \phi) + \dots, \quad (4)$$

where  $k_F = \frac{\pi}{2}(1 - M)$ ,  $\alpha$  and  $\beta$  are constants, and the ellipses indicate higher harmonics.<sup>16</sup>

The main contribution in the low-energy limit comes from the constructive interference between the modulation term  $A_1 \delta_M(x) + A_2 \delta_M(x)^2$  and the most relevant part of  $\rho(x)$ , i.e.,  $\cos(2k_F x + \sqrt{2\pi} \phi)$ . This operator has conformal dimension that depends on the Tomonaga-Luttinger parameter  $K(M, \Delta)/2$ , where  $\Delta$  measures the  $z$ -axis anisotropy in the  $XXZ$  model. Here we emphasize the dependence on the magnetization  $M$  and the anisotropy  $\Delta$ .

Let us propose a periodic pattern of deformations  $\delta_M(x)$  with period  $L_p$ , i.e., satisfying  $\delta_M(x + L_p) = \delta_M(x)$  (the lattice spacing  $a$  is set to 1 in what follows, so that  $L_p$  is an integer). The most general ansatz for the modulation term is given by

$$\delta_M(x) = \sum_{n=1}^{N_w} \delta_n(M) \cos\left(n \frac{2\pi x}{L_p} + \theta_n(M)\right), \quad (5)$$

where  $\delta_n(M)$  are the amplitudes and  $\theta_n(M)$  the phases of the different terms in the expansion. The upper sum index  $N_w$  equals  $L_p/2$  if  $L_p$  is even and  $(L_p - 1)/2$  if it is odd. [In what follows the dependence of  $\delta_n(M)$  and  $\theta_n(M)$  on  $M$  is suppressed to ease the notation, i.e.,  $\delta_n(M) \rightarrow \delta_n$  and  $\theta_n(M) \rightarrow \theta_n$ .]

From Eqs. (4) and (5), we see that the product between the two terms is commensurate whenever the following relation is satisfied:

$$k_F \propto \frac{2\pi}{L_p}, \quad (6)$$

which implies that the wavelengths of the modulations that could pin the relevant cosine term are related to the magnetization as

$$L_p = \frac{4m}{1 - M}, \quad (7)$$

where  $M \neq 1$  and  $m$  is an arbitrary integer—the smallest possible that makes  $L_p$  an integer.

The ansatz in Eq. (5) is verified *a posteriori* from the Density Matrix Renormalization Group (DMRG) analysis, where it is seen that the modulation amplitudes  $\delta_n$  and the phases  $\theta_n$  depend strongly on the value of the magnetization  $M$ , some of them being zero in certain cases.

Using this form for  $\delta_M(x)$ , the interaction term (3) takes the form

$$H_{\text{sp-ph}} = \sum_{p=0}^{2(N_w+1)} \lambda_p \int dx \cos(p k_F x + \sqrt{2\pi} \phi + \Gamma_p), \quad (8)$$

where  $\Gamma_p$  is a function of the phases  $\theta_n$  in the expansion (5) and  $\lambda_p$  is a function of  $\delta_n$ ,  $\theta_n$ , and the coupling constants  $A_1$  and  $A_2$ .

This form of the interaction allows us to conclude that the spin Peierls effect takes place in the usual manner (see Ref. 15 and references therein), since we have both the always commensurate term ( $p = 0$  in the above equation)  $\cos(\sqrt{2\pi} \phi)$  and the  $4k_F$  term that provide together a dimerization of the lattice and a plateau at  $M = 0$  in the magnetization curve.

For finite magnetization, using Eqs. (6)–(8) and using the commensurability condition that arises from Eq. (8),  $p k_F / 2\pi \in \mathbb{Z}$ , one obtains the following condition for the frequencies in Eq. (5) to pin a relevant perturbation:

$$(z \pm 2)(1 - M) = 4 \times \text{integer}, \quad (9)$$

where  $z$  is an integer that runs through all the frequencies that appear in the lattice deformation Eq. (5) and its square, i.e.,  $z = 0, \dots, 2N_w$ . In Table I, we show some examples that we analyze in what follows using DMRG.

One should stress that, in the present case, the situation is rather different than in previous studies of spin systems in a magnetic field, such as in the case of spin ladders, magnetoelastic zig-zag chains, etc., since now the perturbing operator that would be responsible for the plateau is relevant, independently of the values of the microscopic parameters. This may seem to imply that condition (9) is also sufficient, but bosonization alone does not provide the actual values of the amplitudes of the different Fourier components of the

TABLE I. Possible frequencies for the lattice deformations for magnetizations,  $M = 1/5$ ,  $1/3$ , and  $1/2$ , obtained from Eq. (9).

	$M = 1/5$	$M = 1/3$	$M = 1/2$
$L_p$	5	6	8
$N_w$	2	3	4
$z \pm 2$	5, 10, ...	6, 12, ...	8, 16, ...
$z$	3	4	6
Possible frequencies	$k_F$ and $2k_F$	$2k_F$ or $k_F$ and $3k_F$	$2k_F$ and $4k_F$

deformation we proposed and it remains to be checked that they are indeed nonvanishing. In order to answer this question, we need to use DMRG as we describe below.

From the above analysis, we predict that the magnetization curve may present new features related to the frequencies that appear in the Fourier decomposition of the elastic deformation (5) for some given values of  $M$ , such as  $M = 1/5$ ,  $M = 1/3$ ,  $M = 1/2$ , etc. Since these frequencies pin the very relevant term  $\cos(\sqrt{2\pi}\phi)$ , plateaux at these values of  $M$  are expected to show up even for a small anharmonicity  $A_2$ . In such cases, the plateaux widths  $\text{gap}(M, \Delta)$  should scale as

$$\text{gap}(M, \Delta) \propto \lambda^{1/[2-d(M, \Delta)]}, \quad (10)$$

where  $\lambda$  is the coupling constant associated to the relevant cosine term in  $H_{\text{sp-ph}}$  and  $d(M, \Delta)$  is the scaling dimension that can be computed from the Bethe ansatz solution.<sup>17</sup> The coupling  $\lambda$  is a function of the anharmonic amplitude  $A_2$  and its functional dependence, though not predicted by bosonization, can be computed numerically as we show below. From now on, we will concentrate in the isotropic case  $\Delta = 1$ .

#### IV. DMRG ANALYSIS

This is the general setting obtained from bosonization, which provides the qualitative picture expected when anharmonic effects play a role. To have a complete and more quantitative picture, we study the system using extensive DMRG computations. More specifically, we compute the ground-state energy  $E(S_{\text{total}}^z, h = 0)$  of Eq. (1) in the complete set of  $S_{\text{total}}^z$  subspaces using periodic boundary conditions, and keeping just 300 states was shown to be enough to assure the accuracy of the calculation. As usual, adding the Zeeman term, we solve the equation  $E(S_{\text{total}}^z, h) = E(S_{\text{total}}^z + 1, h)$  to

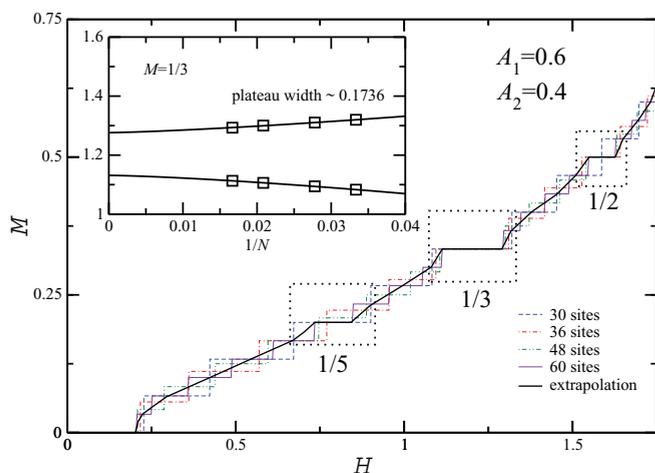


FIG. 1. (Color online)  $M$  vs  $h$  for  $A_1 = 0.6$ ,  $A_2 = 0.4$ , and different system sizes ( $N = 30, 36, 48$ , and  $60$ ). The bold black line corresponds to the extrapolation to the thermodynamic limit. The plateaux at  $M = 0$  and  $1/3$  are clearly observed, while for  $M = 1/5$  and  $1/2$ , it is hard to conclude if they survive in the thermodynamic limit. Note that for  $N = 30$ ,  $M = 1/5$  and  $1/2$  are not commensurate. The inset shows the finite size scaling of the width of the plateau at  $M = 1/3$ . Its finite size scaling is expected to follow  $\text{width}(N) = \text{width}(\infty) + A N^{-B}$ .

obtain the normalized magnetization  $M = 2S_z/N$ , where the plateaux are showing up. This procedure allows us to compute the actual width of the plateaux and their scaling behavior, the deformation patterns, and fractional excitations for the different plateaux.

Let us analyze in detail the situation at  $M = 1/3$ , where we expect to have a plateau. In this case,  $k_F = \pi/3$  and our ansatz for the modulation (5) takes the form

$$\delta_{1/3}(x) = \delta_1 \cos(k_F x + \theta_1) + \delta_2 \cos(2k_F x + \theta_2) + \delta_3 \cos(3k_F x + \theta_3), \quad (11)$$

which leads to the perturbation Hamiltonian

$$H_{\text{sp-ph}} \approx \lambda_{1/3} \cos(\sqrt{2\pi}\phi + \Gamma_{1/3}) + \dots, \quad (12)$$

where  $\lambda_{1/3}$  and  $\Gamma_{1/3}$  depend on  $\lambda_0$  and  $\lambda_6$ , which are the only two commensurate terms in Eq. (8) at magnetization  $M = 1/3$  (see Fig. 1). The dots in Eq. (12) indicate less relevant terms, which can be safely discarded in the presence of the more relevant term  $\propto \cos(\sqrt{2\pi}\phi)$ .

The couplings appearing in Eq. (12) have a lengthy expression in terms of the strengths of the spin-phonon couplings  $A_1$  and  $A_2$  but also on the  $\delta_n$ 's and on the relative phases  $\theta_n$ 's, whose values cannot be extracted from the bosonization analysis alone. To further proceed, we now resort to the numerical analysis of the system using DMRG on large systems, which allows us to estimate all these parameters in a self-consistent way.

The lattice deformations can be calculated in a self-consistent way. Minimizing the ground-state energy and imposing the following constraint:

$$\sum_j \delta_j = 0, \quad (13)$$

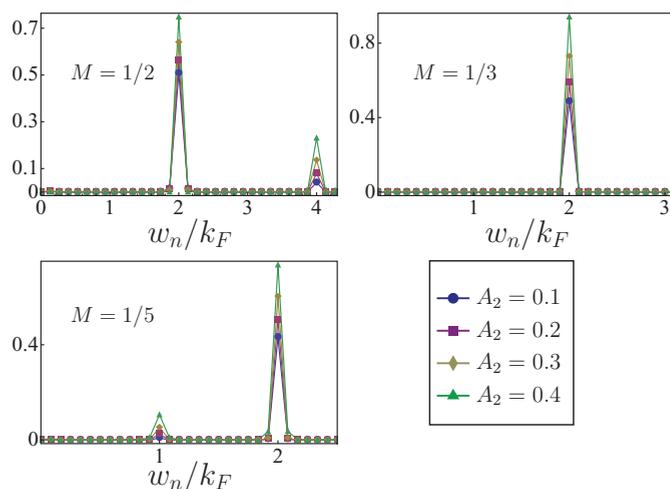


FIG. 2. (Color online) Amplitudes  $\delta_n(M)$  [see Eq. (5)] as a function of the frequency  $w_n = 2\pi n/L_p$  in units of  $k_F$  (with  $A_1 = 0.6$ ) for  $M = 1/2, 1/3$ , and  $1/5$ . The peaks indicate which frequencies contribute to the deformation pattern. The  $2k_F$  peak is always bigger because, to linear order in  $\delta_M(x)$ , it contributes to the energy for all magnetizations.

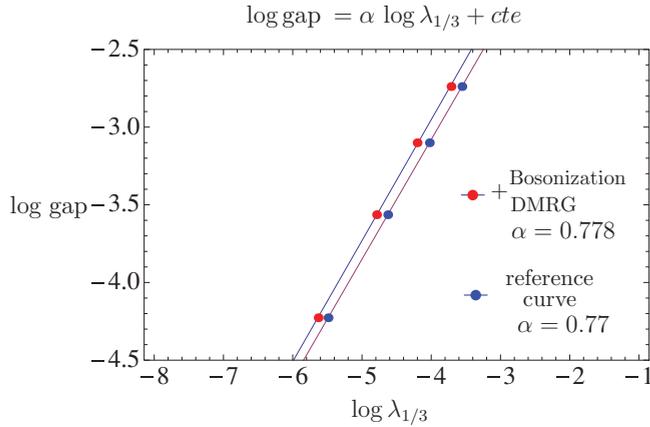


FIG. 3. (Color online) Logarithmic plot of the gap( $\lambda$ ): the blue dots correspond to the reference curve  $\text{gap}(\lambda) = \lambda^{0.77}$  with the gap obtained from DMRG, while the red dots correspond to the gap obtained from DMRG vs the values of  $\lambda$  extracted from bosonization. The value 0.77 is obtained from the Bethe ansatz solution.

we obtain

$$\delta_i = \frac{JA_1 \left[ \left( \frac{\sum_k \langle \vec{S}_k \cdot \vec{S}_{k+1} \rangle (\omega_0 + 2JA_2 \langle \vec{S}_k \cdot \vec{S}_{k+1} \rangle)^{-1}}{\sum_k \langle \omega_0 + 2JA_2 \langle \vec{S}_k \cdot \vec{S}_{k+1} \rangle \rangle^{-1}} \right) - \langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle \right]}{(\omega_0 + 2JA_2 \langle \vec{S}_i \cdot \vec{S}_{i+1} \rangle)}. \quad (14)$$

We start from an arbitrary chosen initial set of deformations  $\{\delta^{(0)}\}$  to be varied and determined self-consistently. For a given set  $\{\delta^{(N)}\}$ , we determine the corresponding ground state and then we compute a new set  $\{\delta^{(N+1)}\}$  using Eq. (14), which we use again in the Hamiltonian. Iterating this procedure, we finally obtain a fixed-point configuration of the deformations  $\delta_i^{(N+1)}(\{\delta^{(N)}\}) = \delta_i^{(N)}$ .

From the DMRG data, we observe that for  $M = 1/3$  only the  $2k_F$  mode contributes to the lattice deformations (see Fig. 2), so that we can safely set  $\delta_1 = \delta_3 = 0$ . As for the phase  $\theta_2$ , it is negligible within the numerical precision so we set it to zero in what follows. With this input from DMRG, we get the following expressions for the bosonization parameters, i.e., for the amplitude  $\lambda_{1/3}$  and phase  $\Gamma_{1/3}$  in Eq. (12),

$$\lambda_{1/3} \propto \left( \frac{A_1 \delta_2}{\sqrt{8}} + \frac{A_2 \delta_2^2}{\sqrt{32}} \right), \quad (15)$$

$$\Gamma_{1/3} = -\pi/3.$$

Here a word is in order: To analyze the scaling of the gap, we need to identify the effective coupling constant associated to the perturbing operator responsible for the opening of the gap. Since the term proportional to  $A_1$  is present for all magnetizations, it does not play a role in the gap opening

and we can then identify the coupling constant governing the scaling of the gap in Eq. (10) as  $\lambda \propto A_2 \delta_2^2$ .

On the other hand, we can extract the deformation amplitude as a function of  $A_2$  from the numerical data, which after a finite-size scaling analysis and a square fit leads to  $\delta_2 = a + bA_2 + cA_2^2$ , with  $a = 0.110$ ,  $b = 0.098$ , and  $c = 0.551$ . Now that we have the dependence of the effective coupling  $\lambda_{1/3}$  on the anharmonicity  $A_2$  we can analyze the scaling of the spin gap (the width of the plateau), which should scale as in Eq. (10).

In order to compare both approaches, we need to use the relation (15) between  $\lambda_{1/3}$  and  $A_2$ , together with the values of  $\delta_2$  obtained from DMRG. Following this approach, in Fig. 3 we show a logarithmic plot of the gap vs  $\lambda_{1/3}$  using the values of  $\lambda_{1/3}$  obtained by bosonization and those of the gap by DMRG (red points). We show a linear fit to obtain the exponent in Eq. (10) and compare with a reference line (blue points) to show the agreement of both approaches.

## V. CONCLUSIONS

In conclusion, we have described a new mechanism leading to the formation of rational magnetization plateau phases, which is mainly due to the anharmonic spin-phonon coupling. We have shown that its role is to pin magneto-elastic deformations that are not present in the harmonic case. By means of bosonization, we have shown that the inclusion of the anharmonic spin-phonon coupling gives as a contribution a relevant operator that is responsible for the plateau in the magnetization curve for certain commensurate values of the magnetization  $M$ . We have performed extensive DMRG computations to complement the analytical computations, since the bosonization approach alone does not provide the actual values of the amplitudes of the different Fourier components of the lattice deformations. In particular, we have analyzed in detail the situation at  $M = 1/3$ , where we have computed the plateau width as a function of the anharmonic coupling, to extract the scaling dimension of the relevant operator that opens the gap. Finally, we have seen that the exponent obtained from the DMRG computations and the one obtained from the Bethe ansatz through bosonization are in excellent agreement, providing further support to our results.

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