Magnetization Plateaus Induced by a Coupling to the Lattice

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We present a novel mechanism for the appearance of magnetization plateaus in quasi-one-dimensional quantum spin systems, which is induced by the coupling to the underlying lattice. We investigate in detail a simple model of a frustrated spin-$1/2$ Heisenberg chain coupled to adiabatic phonons under an external magnetic field, but the present mechanism is expected to be more general. Using field theoretic methods complemented by extensive density matrix renormalization group techniques, we show that magnetization plateaus at nontrivial rational values of the magnetization can be stabilized by the lattice coupling. We suggest that such a scenario could be relevant for some low dimensional frustrated spin-Peierls compounds.

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The field of quantum spin systems offers a wonderful playground for both theorists and experimentalists to investigate a variety of exotic phases cooperatively induced by frustration and a magnetic field [1]. In addition, low dimensional spin systems such as spin chains and ladders have revealed very interesting properties such as the presence of plateaus in their magnetization curves [2,3]. It has been shown theoretically that plateaus occur in general at rational fractions of the saturation magnetization [2]. The position of these plateaus is subject to a quantization condition that involves the volume of a translationally invariant unit cell. From the experimental side, different materials have been found which exhibit plateaus [4]. These well understood cases have in common the fact that the appearance of a given plateau is directly connected with the opening of a spin gap.

In this Letter, supported by both analytical and numerical calculations, we argue that a moderate lattice coupling can generate an extremely rich magnetic phase diagram with a zoo of new $M = p/q$ (rational) plateau states. The main difference from the previously studied cases of purely magnetic systems, is that, as we show here, a different mechanism is responsible for the stabilization of a plateau. We show, in particular, that due to the coupling to the lattice distortions which adapts to the spin modulation, a spin gap is always present, which could be measured, e.g., in inelastic neutron experiments, but plateaus appear only at certain special (rational) values of the saturation magnetization.

Experimentally, the lattice coupling is known to be crucial in spin-Peierls materials like $CuGeO_3$ [5]. It has also been proposed to be responsible for a spontaneous tetramerization [6] in the spin-$1/2$ LiV_2O_5 chain compound [7]. A cooperative effect of the magnetic field and the coupling to an adiabatic lattice was shown to produce in two-leg spin ladders long-range modulated structures [8] for several rational values of the magnetization M. Whether these modulated states give rise to magnetization plateaus or not is still an open issue.

The role of the lattice distortions on the stabilization of plateau states has been addressed in [9,10] in connection to the two-dimensional material $SrCu_2(BO_3)_2$ (For *classical* spins on the pyrochlore lattice see [11]).

Such an investigation is performed here in the particular case of the zigzag chain geometry, but the mechanism presented is expected to be more general, whenever adiabatic phonons have to be taken into account. From the experimental point of view, $SrCuO_2$ [12] and copper germanate ($CuGeO_3$) [5] are fairly good experimental realizations of this geometry.

The underlying richness of the zigzag chain physics is manifest, in particular, under an external magnetic field [13,14]. The magnetic phase diagram shows, in particular, a plateau at $M = 1/3$ with spontaneous breaking of the lattice symmetry of period $q = 3$. These two features are expected *simultaneously* from the quantization condition $qS(1 - M))$ integer [2]. It should be stressed that the pure zigzag spin chain model does not show other ($M \neq 0$) plateau phases besides the $1/3$ plateau state. We show below that the situation changes drastically in the presence of lattice distortions.

The Hamiltonian of a frustrated spin chain coupled to adiabatic phonons in a magnetic field (H) is written as,

$$\mathcal{H} = \frac{1}{2}K\sum_i \delta_i^2 + J_1\sum_i (1 - A_1\delta_i)\vec{S}_i \cdot \vec{S}_{i+1} + J_2\sum_i \vec{S}_i \cdot \vec{S}_{i+2} - H\sum_i S_i^z. \quad (1)$$

H is measured in units where $g\mu_B = 1$, δ_i is the distortion of the bond between site i and $i+1$, K the spring constant, and the first term corresponds to the elastic energy loss. J_1 sets the energy scale and we fix $J_1 = 1$ in what follows.

The spin-lattice coupling A_1 is dimensionless so that the distortions δ_i are given in units of the lattice spacing. Following [6] we redefine the coupling strengths as $\tilde{A}_1 = A_1(1/K)^{1/2}$, although the modulations δ_i depend on A_1 and K, separately.

Let us start with the simplest limit $J_2 = 0$. In that case we know that the system dimerizes for any $A_1 \neq 0$, opening a spin gap and hence leading to a plateau at $M = 0$ in the magnetization curve. In the presence of an external magnetic field the lattice distortion adapts to rest commensurate at any value of M.

To recover these well-known results within bosonization, let us construct the bosonized version of (1) for $J_2 = 0$. In the absence of phonons, we can write the low energy Hamiltonian for the spin system as

$$H_{XXZ}^{\mathrm{cont}} = \frac{v}{2}\int dx\left(K_L[\partial_x\tilde{\phi}(x)]^2 + \frac{1}{K_L}[\partial_x\phi(x)]^2\right), \quad (2)$$

where $\tilde{\phi}$ is the field dual to the scalar field ϕ and it is defined in terms of its canonical momentum as $\partial_x\tilde{\phi} = \Pi$. The magnetic field effect enters through the Luttinger parameter K_L and the Fermi velocity v, which depend on the magnetization M.

In the low energy limit, the term $\propto A_1$ gives a contribution to the energy which reads

$$-\beta A_1 \int dx\delta(x)\colon \cos(2k_F x + \sqrt{2\pi}\phi)\colon . \quad (3)$$

It is then straightforward to conclude that the leading instability of the lattice deformation that minimizes the energy takes the form

$$\delta(x) = \delta_0(M)\cos(2k_F x), \quad (4)$$

where $\delta_0(M = 0) = \delta_c$. This corresponds to the so-called fixed modulation which captures the main qualitative features of the model. This statement can be verified by computing the lattice modulations in a self-consistent way following [15], for which a solution can be approximated by (4) plus subleading higher harmonics contributions. The amplitudes of higher harmonics are generically smaller than the leading one and will then not be considered in the following bosonization analysis [16]. At zero field this modulation produces a total energy gain given by

$$E_{\mathrm{mod}}(\{\delta(x)\}) = K\delta_0(0)^2 - \beta A_1\delta_0(0)\int dx\colon\cos(\sqrt{2\pi}\phi)\colon, \quad (5)$$

while for the nonzero field we have

$$\frac{K}{2}\delta_0(M)^2 - \frac{\beta}{2}A_1\delta_0(M)\int dx\colon\cos(\sqrt{2\pi}\phi)\colon - h\frac{M}{2}. \quad (6)$$

If we assume a smooth variation of $\delta_0(M)$ with M [18], we can conclude that we need a finite magnetic field to start increasing M from zero. In that case we have a plateau at $M = 0$ up to a critical field h_c, after which the magnetization jumps to the value M_s such that the product $-h_c M_s/2$ is of the order of the contribution to the energy due to the modulation E_{mod}, Eq. (5). Because of the presence of the relevant term $\propto \cos(\sqrt{2\pi}\phi)$, the system has a spin gap for all magnetizations. However, the situation described above (plateau and jump) occurs only around $M = 0$ and there are no further plateaus in the magnetization curve, in accordance with the numerical results [19]. The ground state structure above the $M = 0$ plateau has been studied extensively (see, e.g., [20] and references therein) and it comes out that a soliton lattice with a periodicity $2k_F$ starts to develop. The only difference found in the magnetization curve between simulations with fixed and adaptive modulation (when the lattice deformation is determined from minimizing the total energy in a self-consistent iterative form) is a change in the order of the transition from $M = 0$, that changes from first to second order.

The presence of a spin gap for all magnetizations *without plateaus* could be measured in inelastic neutron scattering experiments. This can be illustrated by a simple intuitive argument: the time scale of neutrons scattering is too short to lead to a relaxation of the lattice accompanying a spin excitation while the magnetization process is a thermodynamic quantity.

If we add J_2 a different situation can occur, and, in particular, nontrivial plateaus can appear in certain regions of the parameter space. Let us analyze the case of $M = 1/3$ with a modulation of the form $\delta(x) = \delta_0(1/3)\cos(\frac{2\pi}{3}x)$. Combining this modulation with the second harmonics of the energy density $\gamma\colon \cos(4k_F x + 2\sqrt{2\pi}\phi)\colon$ we obtain an interaction energy given by

$$-A_1\delta_0(1/3)\int dx[\beta\colon\cos(\sqrt{2\pi}\phi)\colon + \gamma\colon\cos(2\sqrt{2\pi}\phi)\colon]. \quad (7)$$

To minimize the energy, the second cosine interaction is pinned at the minimum of the first one and hence we have again a particular situation for $M = 1/3$, since the second harmonics becomes commensurate only for this value of the magnetization. The presence of a plateau at $1/3$ depends on the scaling dimension of the second cosine interaction, which depends on J_2, and from a first order analysis one can estimate that it will be relevant for values of J_2 close to the couplings in $CuGeO_3$, in which $J_2 \sim 0.24$–$0.36J_1$.

This is a new generic mechanism for the appearance of a plateau due to the spin-phonon coupling. The novelty is that the plateau is not produced by the commensurability of the main (relevant) harmonics (as for the zero magnetization case) but it is due to the commensurability of the next-to-leading harmonics, whenever it is relevant.

Note that a plateau at $M = 1/3$ is present in the $J_1 - J_2$ chain without phonons, but in that case, the plateau mechanism is the usual one (so-called classical, since it is well visualized in the Ising limit [21]) and it is driven by the operator : $\cos(3\sqrt{2\pi}\phi)$: which needs larger values of J_2/J_1 than in the present case to become relevant. The present situation is thus much more favorable, making it potentially observable in experiments. Moreover, this plateau can be present also in the extreme anisotropic XY case.

To study the transition from and to the plateau at $M = 1/3$, we are in a similar situation as for the $M = 0$ case in the normal chain discussed above from which we conclude that we have jumps in $M(h)$ at both ends of the $M = 1/3$ plateau. It would be interesting to analyze the formation of a soliton lattice similar to that appearing above $M = 0$ in the present case. We expect that the only modification from fixed to adaptive modulations will be again in the order of the transition.

This analysis can be applied to more general situations, e.g., for a single XXZ chain where one can also expect a 1/3 plateau in the Ising regime. In this case one would need a rather big Ising anisotropy $\Delta \gtrsim 10$ for the second harmonics to be relevant [22].

A similar situation is found for $M = 1/2$ for the $J_1 - J_2$ case, where the second cosine in (7) is now replaced by : $\cos(3\sqrt{2\pi}\phi)$: and is hence less relevant. In the present case, a first order estimate hints that the 1/2 plateau could occur at moderate values of J_2. Notice that this third harmonics is responsible for the plateau at 1/3 in the $J_1 - J_2$ case without phonons [13,23].

We now turn to a numerical analysis of the magnetization process of Hamiltonian (1). We have used the density matrix renormalization group (DMRG) method to obtain the ground state energy $E(S_z)$ in each subspace of the S_z operator (the z component of the total spin of the chain) on a finite chain of N_s sites with open (OBC) or periodic boundary conditions (PBC). Furthermore, minimizing $E = E(S_z) - HS_z$ we have found the magnetization $M = \frac{2S_z}{N_s}$ as a function of the applied magnetic field H.

To begin with, we assume a phonon field δ_i with a fixed periodic modulation $\delta_i = \delta_0 \cos[\pi(1 + M)i]$, as in the previous analytic treatment. In Fig. 1 we show the magnetization as a function of H for three different system sizes and open boundary condition. Parameters are $\frac{J_2}{J_1} = 0.4$ (for which no plateau is present in the pure $J_1 - J_2$ chain [24]) and $\tilde{A}_1\delta_0 = 0.4$. A finite size scaling study of the critical fields is shown in the inset of this figure. The plateaus widths at $M = 1/3$ and $M = 1/2$ extrapole to finite (although small) values in the thermodynamical limit, in agreement with the bosonization analysis. Let us proceed in a more general way, assuming periodic boundaries and minimizing the total energy with respect to all nonequivalent lattice coordinates δ_i. We use the iterative procedure proposed by Feiguin *et al.* [15] and implemented within a DMRG approach by Schönfeld *et al.* [25]. The algorithm has been constructed by using an initial (periodic) ansatz for the δ_i parameters and obtaining a new set of δ_i from the adiabatic equation, $\delta_i = \tilde{A}_1\langle \mathbf{S}_i.\mathbf{S}_{i+1}\rangle$, with the constraint $\sum_i \delta_i = 0$. The procedure is iterated until convergence for the energy and the distortions. Obtaining the distortion pattern in all S_z subspaces, the magnetization curve is then generated. In Fig. 2 we show $M(H)$ for $J_2 = 0.5$ and $A_1 = 0.8$. The plateaus at $M = 1/3$ and $M = 1/2$ are clearly seen. A finite size scaling analysis gives 0.1433 for the width of the plateau at $M = 1/3$ and 0.1654 for the one at $M = 1/2$.

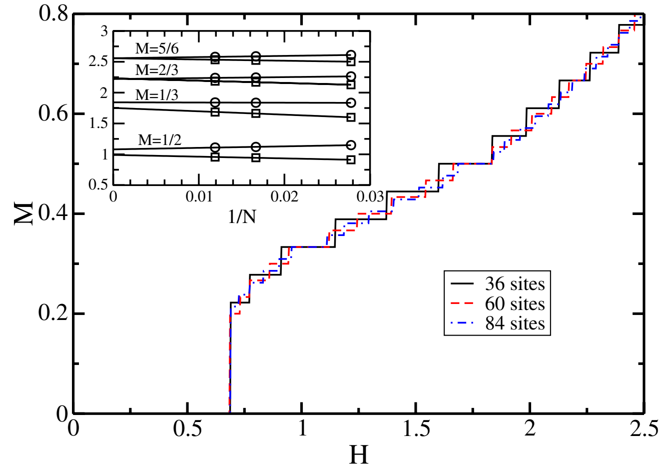

FIG. 1 (color online). $M(H)$ for the 36, 60, and 84 sites for $J_2 = 0.4$ and $\tilde{A}\delta_0 = 0.4$ in the case of fixed modulation and OBC. The inset shows the finite size scaling of the width of the different plateaus.

Finally we performed a careful finite size scaling analysis of the regions of stability of the two most robust plateaus. For that purpose, it is only necessary to consider the S_z values around magnetizations 1/3 and 1/2.

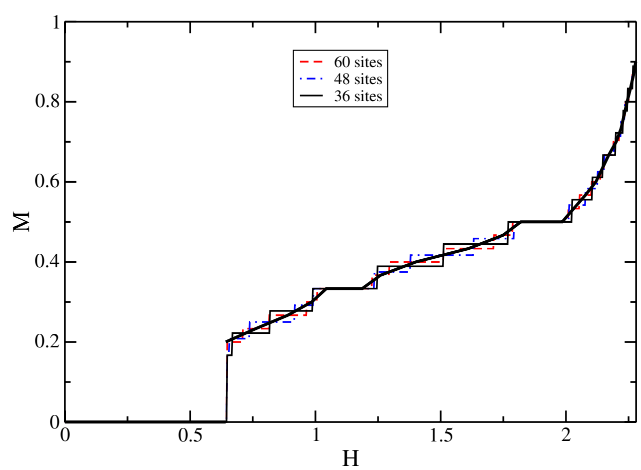

FIG. 2 (color online). $M(H)$ for 36, 48, and 60 sites for $J_2 = 0.5$ and $\tilde{A}_1 = 0.8$ in the case of an adaptive modulation and PBC. The solid line is an extrapolation to the thermodynamical limit.

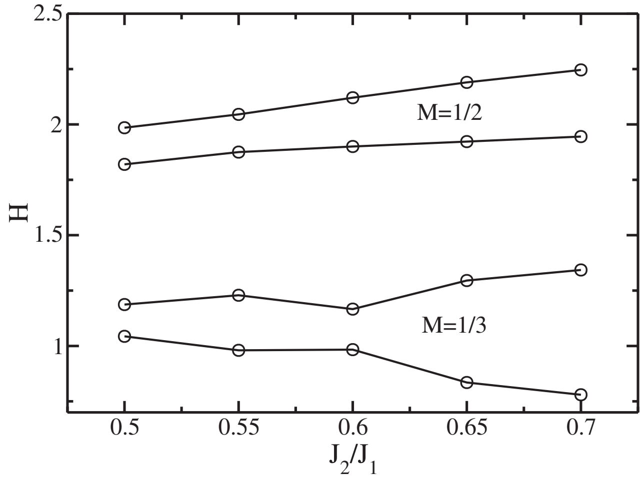

FIG. 3. Region of stability of the $1/3$ and $1/2$ plateaus obtained from a finite size scaling of the critical fields as a function of $\frac{J_2}{J_1}$ in the interval $[0.5, 0.7]$ and for $A_1 = 0.8$. Stability of the other plateau phases is not excluded.

Although we have applied the same iterative procedure as discussed previously, here we have restricted ourselves (at each step) to distortion patterns which fit within the expected supercell [26], a procedure which greatly improves the convergence towards the optimum configuration. The ''phase diagram'' representing the region of stability of the $M = 1/3$ and $M = 1/2$ plateaus with $\frac{J_2}{J_1}$ is shown in Fig. 3. Note that stability of other rational plateau phases suggested by the bosonization approach or by the naive fixed modulation calculation (see Fig. 1) are not at all excluded. However, such phases, which probably have quite narrow widths, are difficult to identify on small clusters.

In conclusion, we have described a new mechanism leading to the formation of rational magnetization plateau phases. It involves a subtle interplay between magnetic frustration and lattice coupling. Our claims are supported by both analytical and numerical calculations. We suggest that a quasi-one-dimensional spin-Peierls systems, like $CuGeO_3$ [5] and tetrathiafulvalene-$AuS_4C_4(CF_3)_4$ [27], where both phonons and frustration play a role, would be the most natural candidates to observe such a phenomenon.

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