Quantum lattice fluctuations in a frustrated Heisenberg spin-Peierls chain

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As a simple model for spin-Peierls systems we study a frustrated Heisenberg chain coupled to optical phonons. In view of the anorganic spin-Peierls compound $CuGeO_3$ we consider two different mechanisms of spin-phonon coupling. Combining variational concepts in the adiabatic regime and perturbation theory in the antiadiabatic regime we derive effective spin Hamiltonians which cover the dynamical effect of phonons in an approximate way. Ground-state phase diagrams of these models are determined, and the effect of frustration is discussed. Comparing the properties of the ground state and low-lying excitations with exact diagonalization data for the full quantum spin-phonon models, good agreement is found especially in the antiadiabatic regime. [S0163-1829(99)04733-5]

I. INTRODUCTION

The effect of a Peierls instability in quasi-one-dimensional spin systems, i.e., the instability of a uniform spin chain towards dimerization induced by the interaction with lattice degrees of freedom, has attracted considerable attention over the last decades. Starting in the 1970s with organic compounds of the TTF and TCNQ family,[1] the interest in the Peierls instability was renewed with the discovery of a spin-Peierls (SP) transition in the anorganic compound $CuGeO_3$ in 1993 by Hase *et al.*[2] The most significant feature distinguishing $CuGeO_3$ from other SP compounds is the high energy of the involved optical phonons, which is comparable to the magnetic exchange integral J. In contrast to the organic materials no softening of these phonon modes is observed near the transition. Therefore the adiabatic treatment of the phonon subsystem used in the works of Pytte[3] or Cross and Fisher,[4] does not seem appropriate to describe the SP transition in $CuGeO_3$, although there are recent efforts in this direction.[5] Rather one has to take into account the effect of quantum lattice fluctuations which tend to decrease the SP transition temperature and the energy gap between the ground state and lowest excitations in the dimerized phase, respectively. Unfortunately there are practically no analytic methods to handle coupled systems of spins (electrons) and phonons when all energy scales and coupling strengths are of the same order of magnitude. This is why many studies involving dynamical phonons rely on numerical methods, such as exact diagonalization (ED),[6,7] density matrix renormalization group (DMRG),[8,9] or Monte Carlo (MC) simulation.[10] Only recently Zheng[11] developed an analytical approach to describe the SP instability of an XY spin chain, which is based on the unitary transformation method. It works well in the adiabatic and antiadiabatic regimes. In the latter case there are also some approaches to the Heisenberg spin chain interacting with optical phonons: Kuboki and Fukuyama[12] used perturbation theory to derive an effective spin Hamiltonian, while Uhrig[13] applied the flow-equation method, i.e., a continuous unitary transformation.[14]

As a simple model which contains all important features of a SP system in the following we consider an antiferromagnetic Heisenberg chain coupled to a set of Einstein oscillators,

$$H = H_s + H_p + H_{sp}, \tag{1}$$

with

$$H_s = J \sum_i \left(\mathbf{S}_i \cdot \mathbf{S}_{i+1} + \alpha \mathbf{S}_i \cdot \mathbf{S}_{i+2}\right), \tag{2}$$

$$H_p = \omega_0 \sum_i b_i^\dagger b_i . \tag{3}$$

The interaction of spins and phonons, H_{sp}, can be modeled in two different ways,

$$H_{sp}^{\mathrm{loc}} = \bar{g} \sum_i \left(b_i^\dagger + b_i\right) \mathbf{S}_i \cdot \mathbf{S}_{i+1}, \tag{4}$$

$$H_{sp}^{\mathrm{diff}} = \bar{g} \sum_i \left(b_i^\dagger + b_i\right) \left(\mathbf{S}_i \cdot \mathbf{S}_{i+1} - \mathbf{S}_i \cdot \mathbf{S}_{i-1}\right), \tag{5}$$

where $\mathbf{S}_i$ denote spin-$\frac{1}{2}$ operators at lattice site i, while $b_i^\dagger$ and b_i are phonon creation and annihilation operators, respectively. H_{sp}^{loc} and H_{sp}^{diff} differ in the mechanism of how the lattice influences the exchange integral. For H_{sp}^{loc}, the *local* coupling, one can think of a single harmonic degree of freedom directly modifying the magnetic interaction. In the context of $CuGeO_3$ this could correspond to side group effects (by the germanium atoms) as discussed in Refs. 15 and 16. In the case of H_{sp}^{diff}, the *difference* coupling, the exchange

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depends directly on the spatial distance between neighboring spins. Note that it is not possible to uniformly decrease or increase all exchange integrals with this type of spin-phonon interaction.

Although H_{sp}^{loc} seems to be more appropriate for $CuGeO_3$ we will consider both variants and compare their properties. In addition we take into account a frustrating next-nearest-neighbor interaction $J\alpha$, which in view of $CuGeO_3$ was introduced to explain susceptibility data.[17,18] As we will see below, the spin-phonon interaction is able to induce this kind of long ranged exchange as well.

Motivated by the success of methods combining unitary transformations with variational and numerical techniques, which we used to study the Peierls transition in the Holstein model of spinless fermions,[19] and inspired by the work of Zheng,[11] in this article we analyze the model (1) within the same framework. In particular we focus on the ground-state phase diagram as a function of spin-phonon coupling, frustration, and phonon frequency, and compare our results with exact diagonalization data.

II. EFFECTIVE SPIN MODELS

To describe a static lattice dimerization in the adiabatic case of small phonon frequency ω_0 we start with a unitary transformation of H which shifts the equilibrium position of each oscillator by a constant amount alternating from site to site, $\tilde{H}=\exp(S_1)H\exp(-S_1)$, with

$$S_1=\frac{\Delta_\pi}{2\bar{g}}\sum_i (-1)^i(b_i^\dagger-b_i). \tag{6}$$

For the terms involving phonons this yields

$$\tilde{H}_p=H_p-\omega_0\frac{\Delta_\pi}{2\bar{g}}\sum_i (-1)^i(b_i^\dagger+b_i)+N\omega_0\left(\frac{\Delta_\pi}{2\bar{g}}\right)^2, \tag{7}$$

$$\tilde{H}_{sp}^{\mathrm{loc}}=H_{sp}^{\mathrm{loc}}-\Delta_\pi\sum_i (-1)^i\mathbf{S}_i\cdot\mathbf{S}_{i+1}, \tag{8}$$

$$\tilde{H}_{sp}^{\mathrm{diff}}=H_{sp}^{\mathrm{diff}}-2\Delta_\pi\sum_i (-1)^i\mathbf{S}_i\cdot\mathbf{S}_{i+1}. \tag{9}$$

Δ_π will act as the variational parameter describing the dimerization of the system.

Applying another unitary transformation $\bar{H}=\exp(S_2)\tilde{H}\exp(-S_2)$, we want to decouple spin and phonon degrees of freedom in the antiadiabatic case of large phonon frequency. In analogy to the (incomplete) Lang-Firsov transformation used in Refs. 19 and 11, we choose the ansatz

$$S_2^{\mathrm{loc}}=f\frac{\bar{g}}{\omega_0}\sum_i (b_i^\dagger-b_i)\mathbf{S}_i\cdot\mathbf{S}_{i+1}, \tag{10}$$

$$S_2^{\mathrm{diff}}=f\frac{\bar{g}}{\omega_0}\sum_i (b_i^\dagger-b_i)(\mathbf{S}_i\cdot\mathbf{S}_{i+1}-\mathbf{S}_i\cdot\mathbf{S}_{i-1}). \tag{11}$$

The free parameter f accounts for the change from the antiadiabtic to the adiabtic regime; numerically it is determined such that the contributions of first order in $\bar{g}$ to the Hamiltonian $\bar{H}$ are as small as possible (reminding us of the usual condition for a Schrieffer-Wolff transformation[20]). In particular we require the amplitude of the state resulting from the application of $\tilde{H}_{sp}+[S_2,\tilde{H}_s+\tilde{H}_p]|_{\Delta_\pi=0}$ to the ground state of H_s+H_p to be minimal. Figure 1 illustrates the variation of f with varying phonon frequency ω_0.

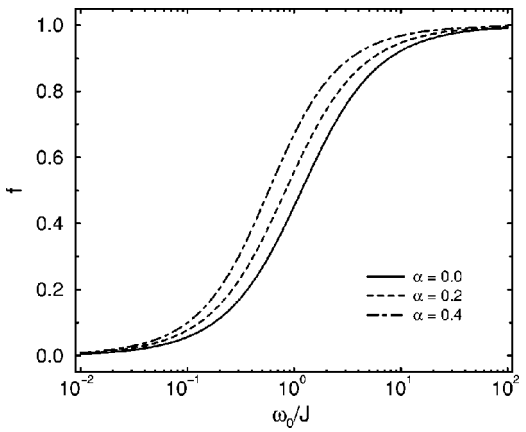

FIG. 1. Variation of f in the case of local coupling and lattice size $N=16$.

We find that the general shape of $f=f(\omega_0)$ depends only weakly on both system size N and frustration α. While $f\to 1$ in the antiadiabatic frequency range ($\omega_0\gg J$), the transformation $\exp(S_2)$ vanishes completely as the frequency becomes small ($\omega_0\ll J$).

In contrast to electron-phonon systems with Holstein coupling, where a transformation similar to $\exp(S_2)$ can be evaluated to give a simple analytic expression, applying the unitary transformation $\exp(S_2)$ to $\tilde{H}$, we obtain an infinite series of terms, which cannot be summed up easily, i.e.,

$$\bar{H}=\sum_k [S_2,\tilde{H}]_k/k!, \tag{12}$$

where $[S_2,\tilde{H}]_k$ denotes the iterated commutator $[S_2,\tilde{H}]_{k+1}=[S_2,[S_2,\tilde{H}]_k]$ with $[S_2,\tilde{H}]_0=\tilde{H}$. In the following, for $\bar{H}$ we will consider only contributions up to fourth order in $\bar{g}$.

To determine properties of the low-energy spin excitation spectrum (like the existence of a spin gap) we have to keep most details of the spin system, and we therefore derive an effective *spin* model by taking the average over the phonon subsystem, $H_{\mathrm{eff}}=\langle\bar{H}\rangle$. As we stay close to the ground state, $\langle\cdots\rangle$ should denote averaging over the phonon vacuum. However, to allow for a later comparison with the results of Uhrig,[13] in an intermediate step we write $\langle\cdots\rangle$ for the average over thermally excited phonon states, and introduce

$$Y:=\langle(b_i^\dagger-b_i)^2\rangle=\begin{cases}-1 & \text{if } T=0,\\ -\coth\left(\dfrac{\omega_0}{2T}\right) & \text{if } T>0,\end{cases} \tag{13}$$

as a shorthand notation for an expression occurring repeatedly below. The resulting spin Hamiltonian $H_{\mathrm{eff}}=\langle\bar{H}\rangle$ contains long-ranged Heisenberg interactions as well as numer-

ous four- and six-spin couplings of the form $(\mathbf{S}_i\cdot\mathbf{S}_j)(\mathbf{S}_k\cdot\mathbf{S}_l)\cdots(\mathbf{S}_m\cdot\mathbf{S}_n)$. To a good approximation we can neglect them and obtain

$$H_{\rm eff}/J = J_0 + \sum_i \left[J_1 + (-1)^i\delta\right]\mathbf{S}_i\cdot\mathbf{S}_{i+1} + \sum_i \sum_{k=2}^{4} J_k \mathbf{S}_i\cdot\mathbf{S}_{i+k}. \quad (14)$$

Note that all phonon dynamics disappeared from the Hamiltonian $H_{\rm eff}$, but the effect of the spin-phonon interaction enters through both the static dimerization parameter Δ_π and the different long-range spin interactions.

For the local coupling the corresponding interaction strengths are

$$\delta^{\rm loc} = -\frac{\Delta_\pi}{J}\left[(1-f)+\frac{f^2g^2Y}{2}\left(1-\frac{f}{3}\right)\right], \quad (15)$$

$$J_0^{\rm loc} = N\left[\frac{\omega_0}{J}\langle b_i^\dagger b_i\rangle + \frac{1}{4\lambda}\left(\frac{\Delta_\pi}{J}\right)^2 - \frac{3}{8}\lambda f\left(1-\frac{f}{2}\right) - \frac{\lambda g^2 Y f^3}{16}\left(1-\frac{f}{4}\right)\right], \quad (16)$$

$$J_1^{\rm loc} = 1+\lambda f\left(1-\frac{f}{2}\right)+\frac{f^2g^2Y(1-\alpha)}{2} + \frac{\lambda g^2 Y f^3}{2}\left(1-\frac{f}{4}\right)+\frac{f^4g^4Y^2}{96}(28-37\alpha), \quad (17)$$

$$J_2^{\rm loc} = \alpha - \frac{f^2g^2Y(1-2\alpha)}{2} - \frac{\lambda g^2 Y f^3}{4}\left(1-\frac{f}{4}\right) - f^4g^4Y^2\frac{37}{96}(1-2\alpha), \quad (18)$$

$$J_3^{\rm loc} = -\frac{f^2g^2Y\alpha}{2}+\frac{f^4g^4Y^2}{96}(9-46\alpha), \quad (19)$$

$$J_4^{\rm loc} = \frac{9f^4g^4Y^2\alpha}{96}, \quad (20)$$

while for the difference coupling we find

$$\delta^{\rm diff} = 2\,\delta^{\rm loc}, \quad (21)$$

$$J_0^{\rm diff} = N\left[\frac{\omega_0}{J}\langle b_i^\dagger b_i\rangle + \frac{1}{4\lambda}\left(\frac{\Delta_\pi}{J}\right)^2 - \frac{3}{4}\lambda f\left(1-\frac{f}{2}\right) - \frac{3\lambda g^2 Y f^3}{16}\left(1-\frac{f}{4}\right)\right], \quad (22)$$

$$J_1^{\rm diff} = 1+2\lambda f\left(1-\frac{f}{2}\right)+\frac{3f^2g^2Y(1-\alpha)}{2} + \frac{3\lambda g^2 Y f^3}{2}\left(1-\frac{f}{4}\right)+\frac{f^4g^4Y^2}{24}(59-75\alpha), \quad (23)$$

$$J_2^{\rm diff} = \alpha+\lambda f\left(1-\frac{f}{2}\right)-\frac{f^2g^2Y(3-5\alpha)}{2} - \frac{\lambda g^2 Y f^3}{12}\left(1-\frac{f}{4}\right)-\frac{f^4g^4Y^2}{24}(75-124\alpha), \quad (24)$$

$$J_3^{\rm diff} = -f^2g^2Y\alpha - \frac{5\lambda g^2 Y f^3}{6}\left(1-\frac{f}{4}\right)+\frac{f^4g^4Y^2}{48}(32-119\alpha), \quad (25)$$

$$J_4^{\rm diff} = \frac{21f^4g^4Y^2\alpha}{48}. \quad (26)$$

To point out the relevant model parameters we introduced the dimensionless coupling constants $\lambda=\bar{g}^2/(J\omega_0)$ (cf. Refs. 3 and 4) and $g=\bar{g}/\omega_0$.

Comparing our result with that of Uhrig,[13] in $H_{\rm eff}^{\rm diff}$ we have to set $f=1$, which corresponds to the antiadiabatic regime, and $\Delta_\pi=0$. Indeed we recover [except for a prefactor 1/2 which in Ref. 13 enters erroneously going from Eq. (11c) to Eq. (13)] all second-order terms derived with the flow-equation method, supplemented by some new fourth-order contributions.

Hereafter we set $T=0$ and use $Y=-1$ and $\langle b_i^\dagger b_i\rangle=0$ exclusively; i.e., we search for a good approximation to the ground state of Eq. (1).

III. TRANSITION TO A GAPPED PHASE

A prominent feature associated with the SP instability is of course the existence of an energy gap between the ground state and lowest excitations. Considering, in a first step, the pure spin model H_s, it is known that the spectrum is gapless for the Heisenberg chain with $\alpha=0$, where the lowest spinon excitations (triplet and singlet) are degenerate with the ground state at momenta $q=0$ and π.[21–23] In contrast the system has a twofold-degenerate ground state and a gap to lowest triplet excitations at $\alpha=0.5$, the Majumdar-Ghosh point.[24] At some intermediate frustration α_c the model undergoes a transition from the gapless to the gapped phase, which is of Kosterlitz-Thouless type.[25–27] Using arguments of conformal field theory one can show that the lowest singlet and triplet excitations of a finite system of size N become degenerate at $\alpha_c(N)$, where the dependence on N is only weak and $\alpha_c(N)-\alpha_c(\infty)\sim N^{-2}$ (cf. Refs. 28–30). This was used in Refs. 31, 18 and 32 to determine $\alpha_c=0.241167(5)$.

Looking at our effective spin models $H_{\rm eff}$ we find that the interaction with the optical phonons induces the same kind of frustrating next-nearest neighbor interaction. Therefore, without any explicit frustration α, the effective frustration $\alpha_{\rm eff}:=J_2/J_1$ due to the phonons can lead to a gap in the energy spectrum and to spontaneous dimerization, as was already discussed in Refs. 12 and 13. This effect is most important in the antiadiabatic frequency range.

Another mechanism producing a gap is (static) dimerization, i.e., an alternation of the nearest-neighbor exchange integral. Taking the adiabatic limit of our effective model, $f\to 0$ and $\delta\to\Delta_\pi$, the ground-state energy of $H_s+\delta\Sigma_i(-1)^i\mathbf{S}_i\cdot\mathbf{S}_{i+1}$ is known to deviate from its value at

$\delta=0$ like $\delta^{4/3}$ (cf. Ref. 4), while the elastic energy increases with δ^2. Therefore for all couplings λ the ground-state energy of $H_{\rm eff}(f=0)$ is minimal, if δ is finite. At the same time proportional to $\delta^{2/3}$ a gap opens in the spectrum.

By taking into account both mechanisms we can now determine the transition from the gapless phase to the gapped one. As the SP system behaves differently for the two couplings, we treat them separately, starting with the *local* coupling case.

A. Local coupling

In a first step we set $\Delta_\pi=0$ and use the level-crossing criterium[31,18,9] to calculate the critical line in the α-g plane for different phonon frequencies ω_0 and system sizes N. Since $H_{\rm eff}^{\rm loc}$ contains longer-ranged interactions such as $\mathbf{S}_i\cdot\mathbf{S}_{i+3}$ and $\mathbf{S}_i\cdot\mathbf{S}_{i+4}$, this line slightly deviates from the line $\alpha_{\rm eff}=\alpha_c$, and we have to calculate it separately. Applying the Lanczos algorithm to the effective model we obtain the critical line with high accuracy on local workstations ($N\leqslant 20$).

On the other hand we determine the level crossing in the original model (1) by using the methods described in Ref. 6. In the case $\omega_0/J=0.1$, the latter is complicated with our Lanczos diagonalization code, since for the small systems we can handle the finite-size gap to that singlet excitation we have to consider for the level crossing is a few times larger than ω_0. Therefore this singlet is in between a number of other singlet excitations, not allowed for determining the critical coupling. Only very large systems (see Ref. 9, Table II) with a finite-size gap smaller than ω_0 permit one to precisely locate the level crossing.

Figures 2(a) and 2(b) show the critical lines in the effective (bold solid) and the original model (bold dashed) as well as the lines $\alpha_{\rm eff}=\alpha_c$ (thin solid). As in the case of the pure spin model, the critical lines depend only weakly on N. We can therefore compare exact data for the original model and $N=8$ with data for the effective model and $N=16$. While the results differ noticeably for intermediate phonon frequency $\omega_0\sim 1$, the agreement is excellent in the antiadiabatic frequency range $\omega_0\gg J$. With increasing ω_0 the critical curve exhibits a remarkable upturn before crossing the abscissa; i.e., the frustration is suppressed for small spin phonon coupling, but overcritical for strong coupling. It is this feature which makes it necessary to expand Eq. (12) up to fourth order to approximate $H^{\rm loc}$ in a correct way. A second-order theory is not capable of describing the observed critical line.

Another point we can study within our effective model is the behavior of the critical spin phonon coupling g_c at $\alpha=0$ in the limit $\omega_0/J\to\infty$, i.e., the limit of the crossing point of the critical line and the abscissa. As the effects of the longer-ranged interactions are rather small, we can solve the equation $\alpha_{\rm eff}^{\rm loc}=\alpha_c$. Setting $f=1$ (compare Fig. 1) and $\alpha=0$ we find

$$g_c^2=\frac{P}{2Q}+\sqrt{\left(\frac{P}{2Q}\right)^2+\frac{\alpha_c}{Q}}, \tag{27}$$

with

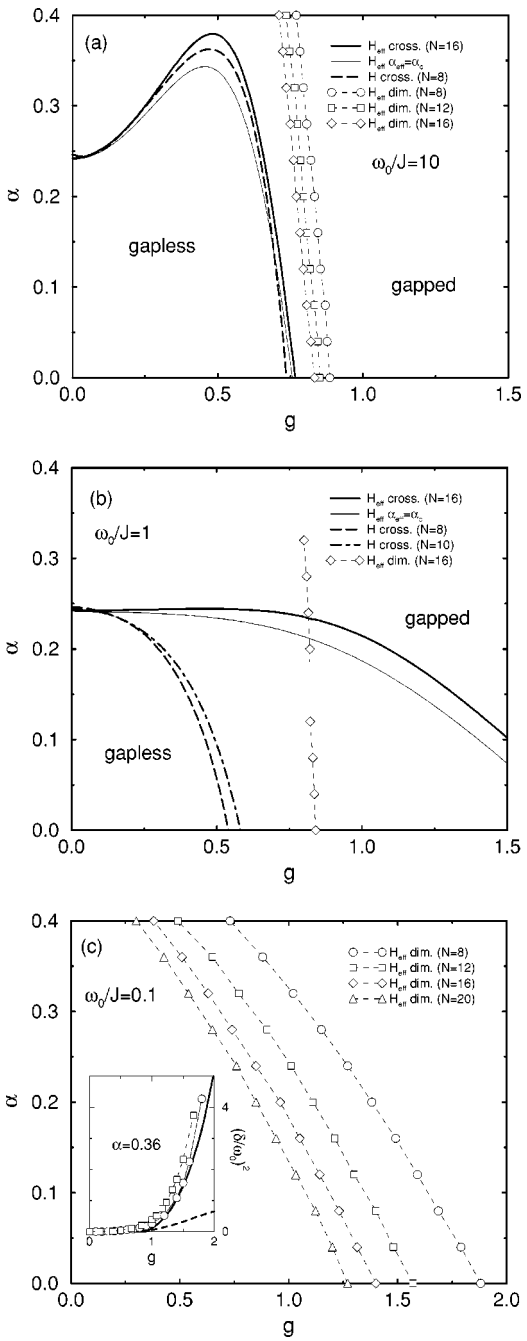

FIG. 2. Singlet-triplet level crossing (solid lines) and onset of dimerization (dashed lines with symbols) in the effective model in comparison to the level crossing in the original model (bold dashed lines) at $\omega_0/J=10$, 1 and 0.1. In (c) the inset shows $(\delta/\omega_0)^2$ for $\alpha=0.36$, $\omega_0/J=0.1$ (solid line), 0.316 (dashed line), and $N=8$.

$$P=\frac{\omega_0}{J}\frac{\alpha_c}{2}-\frac{1}{2}(1+\alpha_c), \tag{28}$$

$$Q=\frac{\omega_0}{J}\frac{3}{8}\left(\frac{1}{2}+\alpha_c\right)-\left(\frac{37}{96}+\frac{7}{24}\alpha_c\right), \tag{29}$$

and in the limit of infinite phonon frequency g_c approaches a finite value,

$$\lim_{\omega_0/J\to\infty} g_c=\sqrt{\frac{8\alpha_c}{3(1+2\alpha_c)}}\approx 0.66, \tag{30}$$

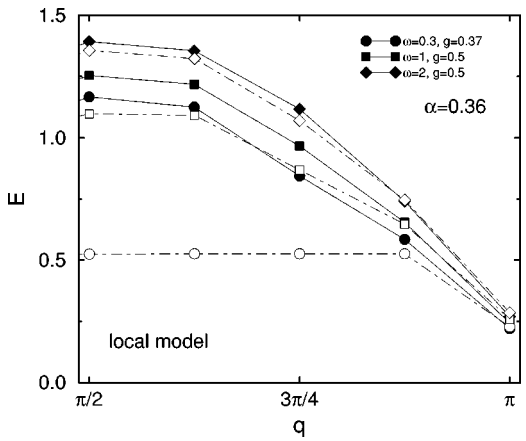

FIG. 3. Low-lying excitations in the effective (solid symbols) and the original model (open symbols) for different frequency and local coupling g.

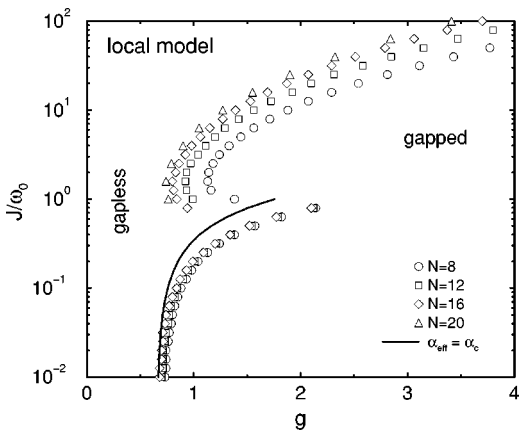

FIG. 4. Critical coupling g_c vs frequency for $H_{\mathrm{eff}}^{\mathrm{loc}}$ with $\alpha=0$.

for the model with local coupling.

In the case small phonon frequency, $\omega_0 \ll J$, the second transformation $\exp(S_2)$ loses its importance, and the effective frustration due to the spin-phonon interaction is replaced by the dimerization as the relevant mechanism leading to an energy gap. We account for this effect by allowing for a finite Δ_π in our approximation. Using the Hellmann-Feynman theorem and numerical diagonalization of finite-spin systems we determine Δ_π such that the ground-state energy of $H_{\mathrm{eff}}^{\mathrm{loc}}$ is minimal. Depending on coupling strength g and frustration α the system prefers to remain in the undimerized, gapless phase ($\Delta_\pi=0$) or to develop a nonzero dimerization leading to a gap. In Figs. 2(a)–2(c) we plotted these transition lines (dashed line with symbols) in addition to those obtained by level crossing. As we already found in our study of the Holstein model of spinless fermions,[19] for small ω_0 the transition to the dimerized phase depends noticeably on the system size N [see Fig. 2(c)], while the finite-size dependence is weak in the antiadiabatic regime [cf. Fig. 2(a)]. In addition, for $\omega_0/J=10$ the transition is consistent with the critical line determined via level crossing.

To compare properties of the original and effective models also in the case of small phonon frequency, we consider the dimerization. As a quantity which corresponds to $\delta^{\mathrm{loc/diff}}$ we take the static (lattice) structure factor,[6,7]

$$\delta^2=\frac{\bar{g}^2}{N^2}\sum_{j,k}\langle u_j u_k\rangle e^{i\pi(j-k)}, \tag{31}$$

calculated in the ground state of Eq. (1), where $u_j=b_j^\dagger+b_j$. The inset of Fig. 2(c) demonstrates that the results for the dimerization in the effective (bold lines) and the original model (thin lines with symbols) agree rather well, especially for $\omega_0/J=0.1$.

Another feature we can compare is the dispersion of low-lying excitations. Figure 3 shows the energy of the lowest triplet excitations, calculated exactly and within our approximation. Clearly for $\omega_0/J=0.3$ the correct dispersion is flattened at momenta near $q=\pi/2$. This results from the energy of the dispersionless phonons, which is added to the lowest triplet at $q=\pi$. Of course our effective model does not contain these low-lying phonon excitations. However, as soon as $\omega_0 \gtrsim J$ the lowest excitations are due to renormalized spin interactions and well approximated by the effective model.

To collect the results of this subsection we show in Fig. 4 the critical coupling $g_c(\alpha=0)$ over a wide range of phonon frequencies, using both criteria for the phase transition. Symbols stand for the onset of dimerization, while the bold line corresponds to $\alpha_{\mathrm{eff}}=\alpha_c$. As expected, we find that our approximation is somewhat unreliable for intermediate phonon frequencies. The singularity of g_c at $\omega_0/J=1$ is a manifestation of this deficiency. The correct critical line will connect adiabatic and antiadiabatic behavior in a continuous way (compare also the next subsection and Ref. 9).

B. Difference coupling

The procedure to determine the phase transition in the SP system with difference coupling is the same as described before. In the antiadiabatic regime we set $\Delta_\pi=0$ and calculate the position of the crossing of the first triplet and the first singlet excitation for both the original and effective models. The results for $\omega_0/J=10$ and 1 are shown in Figs. 5(a) and 5(b), respectively.

In contrast to the local coupling the structure of the critical line for high phonon frequency ($\omega_0/J=10$) is much simpler. It appears that one would get the same shape also for a second-order theory. However, to enlarge the application area of our approximation taking into account higher-order contributions is still appropriate. As before, the agreement between the original and effective models is excellent in the antiadiabatic regime, while the deviations increase with approaching intermediate frequencies.

Calculating the behavior of the critical coupling $g_c(\alpha=0)$ in the limit of infinite phonon frequency, $\omega_0/J\to\infty$, we now find

$$g_c^2=-\frac{P}{2Q}+\sqrt{\left(\frac{P}{2Q}\right)^2+\frac{\alpha_c}{Q}}, \tag{32}$$

with

$$P=\frac{\omega_0}{J}\left(\frac{1}{2}-\alpha_c\right)+\frac{3}{2}(1+\alpha_c), \tag{33}$$

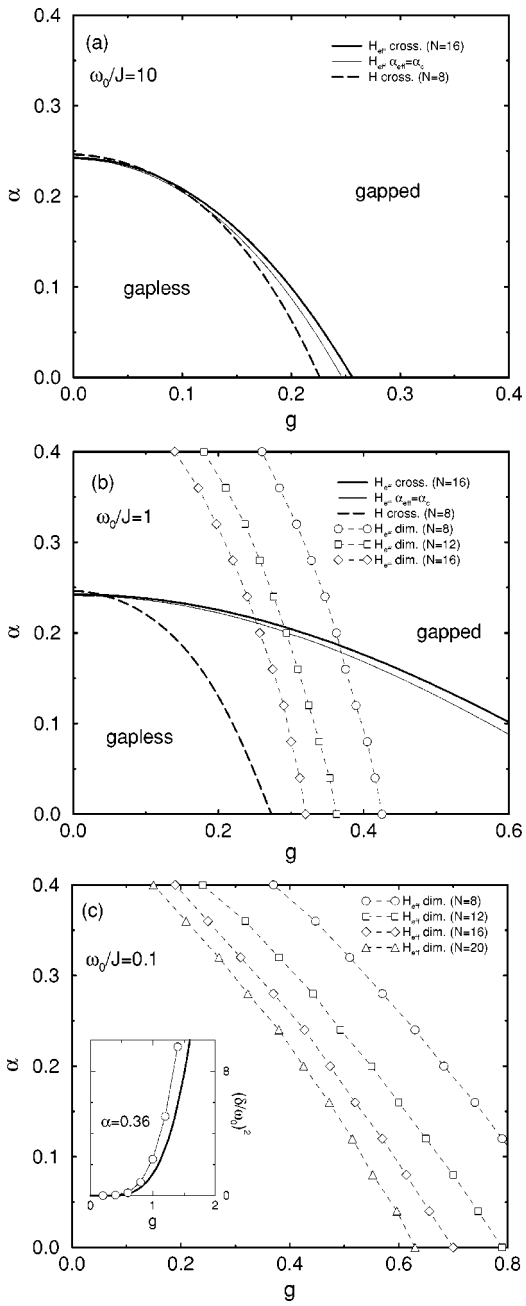

FIG. 5. Level crossing (solid lines) and onset of dimerization (dashed lines with symbols) in the effective model in comparison to the level crossing in the original model (bold dashed lines) at $\omega_0/J=10$, 1 and 0.1. In (c) the inset compares exact (symbols) and variational results for $(\delta/\omega_0)^2$ at $\alpha=0.36$, $\omega_0/J=0.1$, and $N=8$.

$$Q=\frac{\omega_0}{J}\left(\frac{1}{16}+\frac{9}{8}\alpha_c\right)-\left(\frac{25}{8}+\frac{59}{24}\alpha_c\right), \tag{34}$$

and, differently from the local coupling case, g_c tends to zero,

$$\lim_{\omega_0/J\to\infty} g_c=0. \tag{35}$$

While the $q=0$ and the $q=\pi$ phonon modes compete in the case of local spin phonon coupling, allowing for a stable gapless phase up to a critical g, there is no interaction with

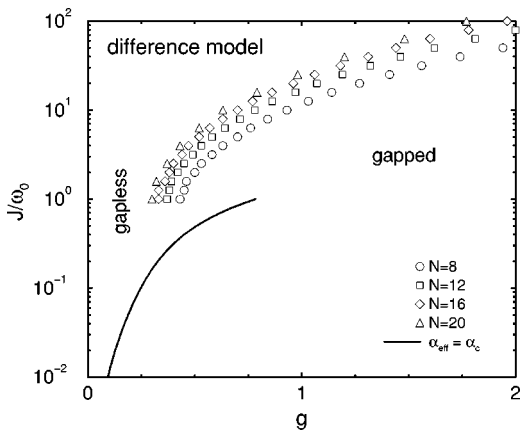

FIG. 6. Critical coupling g_c vs frequency for $H_{\rm eff}^{\rm diff}$ with $\alpha=0$.

the $q=0$ mode in $H_{sp}^{\rm diff}$. Therefore the $q=\pi$ mode induces long-ranged exchange more efficiently, leading to a vanishing g_c for $\omega_0/J\to\infty$.

For small phonon frequencies $\omega_0\ll J$ again we determine the optimal dimerization Δ_π and the critical line beyond which Δ_π starts to be nonzero. The results are shown in Figs. 5(b) and 5(c). For large phonon frequency we find an unstable behavior of Δ_π; it is finite for some g, but vanishes before growing to substantial values again. We therefore make no attempt to fix the onset of $\Delta_\pi\neq 0$ for $\omega_0/J\gtrsim 1$.

As in the case of local coupling the dimerization in the original and effective models agrees well for $\omega_0/J=0.1$ [inset Fig. 5(c)].

Finally in Fig. 6 we combine the above results to obtain the phase diagram in the J/ω_0-g plane. Except for $\omega_0/J\to\infty$, where $g_c\to 0$, the behavior is similar to the case of local coupling. Again at $\omega_0/J\sim 1$ the effective model gives a singularity in g_c, while the correct result should be continuous. Comparing the transition line with the very recent DMRG data of Bursill *et al.*,[9] we find very good agreement in the antiadiabatic regime (the phase transition does not change, going from the second- to fourth-order effective theory). For small phonon frequency $\omega_0/J\to 0$ the results of Bursill *et al.* suggest a finite limit for g_c/ω_0, while our model gives an increasing ratio of g_c/ω_0. However, as the finite-size effects are large for $\omega_0\ll J$, determining the correct value of g_c is a delicate procedure and the exact g_c might be much smaller than depicted in Fig. 6.

IV. GROUND-STATE PHONON DISTRIBUTION

In the course of exact diagonalizations of the phonon dynamical model (1) we observed another interesting feature distinguishing the two mechanisms of spin-phonon interaction. Turning our attention to the phonon distribution in the ground state of Eq. (1), we find for the model with *difference* coupling in the adiabatic regime that the system prefers states with *even* phonon occupation numbers [cf. Fig. 7(a)]. This behavior reminds us of a simple two-level system, more precisely the Rabi (pseudo-Jahn-Teller) Hamiltonian[33]

$$H=\Delta\sigma_z+\bar{g}(b^\dagger+b)\sigma_x+\omega_0 b^\dagger b\ \sigma_0, \tag{36}$$

where an interaction with a bosonic degree of freedom connects the levels. Solving this model in the adiabatic strong-

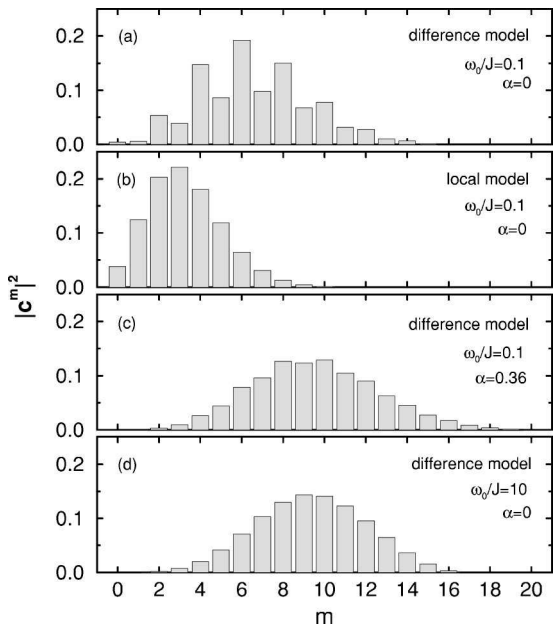

FIG. 7. (a) Phonon distribution in the ground state of the model with difference coupling ($g=1.4$, $\omega_0/J=0.1$) and (b),(c) mechanisms that suppress the even-odd imbalance: local coupling, frustration, and antiadiabaticity.

coupling case $\omega_0/\Delta \ll 1$ and $g=\bar{g}/\omega_0 > 1$, we obtain a similar even-odd alternation in the ground-state phonon distribution. One can understand this effect within standard perturbation theory. Starting with the two levels $\pm\Delta$ and the corresponding eigenstates $\binom{1}{0}$ and $\binom{0}{1}$, it is obvious that adding only *one* phonon requires an extra amount of 2Δ exchange energy, because the system is excited to the upper level. Therefore the system prefers an even number of phonons for the ground-state wave function. In the antiadiabatic case $\omega_0 \gg \Delta$, to a good approximation we can omit the $\Delta\sigma_z$ term and solve the model by unitary transformation, leading to a Poisson distribution of the phonons (coherent state).

In view of the SP system with difference coupling, the $q=\pi$ phonon is the most relevant (in fact in the ground state almost all phonons occupy this mode), and adding one phonon changes the momentum of the spin system by π. Consequently the gap between the (singlet) ground state at $q=0$ ($N=4k$, $k\in\mathbb{N}$) or π ($N=4k+2$) and the lowest singlet state at $q=\pi$ or 0, respectively, plays the role of 2Δ in the two-level system. As this singlet-singlet gap is only due to the finite size of the considered spin chain, we believe that the even-odd alternation will disappear in the thermodynamic limit. The reduction (disappearance) of the even-odd imbalance resulting from a finite frustration with a much smaller singlet-singlet gap (zero at $\alpha=0.5$) [cf. Fig. 7(c)] can be taken as a first indication. Of course we observe a smooth phonon distribution in the antiadiabatic case [Fig. 7(d)].

The model with local spin-phonon coupling exhibits the usual Poisson distribution for all frequencies, since the interaction is due to the $q=\pi$ *and* the $q=0$ phonon mode [Fig. 7(b)].

V. CONCLUSION

In summary, we have studied the spin-Peierls instability of a frustrated Heisenberg spin chain coupled to optical phonons of energy ω_0. Using the concept of unitary transformations we derive effective spin Hamiltonians, which cover the spin-phonon interaction by two mechanisms, static dimerization Δ_π and long-ranged exchange couplings. Both can lead to an energy gap between the ground state and lowest excitations, which is related to a Peierls instability of the spin system. In the antiadiabatic phonon frequency range $\omega_0 \gg J$, we verify and extend the Hamiltonian obtained recently with the flow-equation method,[13] while we recover the usual static SP model in the limit $\omega_0/J \to 0$.

To determine the transition to the gapped phase in the case of large phonon frequency we use the level-crossing criterium, which proved to be very accurate for similar models.[31,18,9] For the two types of spin-phonon coupling [*local*, $u_i\ \mathbf{S}_i\cdot\mathbf{S}_{i+1}$, and *difference*, $(u_i-u_{i+1})\ \mathbf{S}_i\cdot\mathbf{S}_{i+1}$] we consider here, the results of our effective models agree very well with data from exact diagonalization of the original, phonon dynamical model and with recent DMRG data[9] (difference coupling only). In the case of local coupling two phonon modes ($q=0$ and π) compete and allow for a gapless phase to exist in a wider parameter range. Furthermore, we observe a nonmonotonous behavior of the phase transition line as a function of spin-phonon interaction and frustration α. With difference coupling spins and phonons interact almost only through the π mode, which is able to dimerize the system most efficiently.

For phonon frequencies $\omega_0 \lesssim J$, we determine the phase transition by means of the static dimerization Δ_π, which changes from zero at small spin phonon coupling to a finite value beyond a critical coupling.

At intermediate frequency the situation remains unsatisfactory as the critical coupling behaves discontinuously. Here numerical methods, including the full phonon dynamics, still provide the only reliable tool to study the transition. Nevertheless, the proposed effective models help to understand the physical mechanisms leading to spontaneous dimerization of the interacting spin-phonon system.

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