Effect of spin-phonon coupling on the Haldane gap in antiferromagnetic Heisenberg chains

M. E. Gouvêa*
Centro Federal de Educação Tecnológica de Minas Gerais, Belo Horizonte, MG, Brazil

A. S. T. Pires
Departamento de Fisica, ICEx, Universidade Federal de Minas Gerais, Belo Horizonte, MG, Brazil

(Received 6 October 2006; revised manuscript received 19 December 2006; published 5 February 2007)

The effect of the spin-phonon coupling on the spin-1 Heisenberg antiferromagnetic chain is investigated. Using the framework of the modified spin-wave theory, we show that the gap is increased when the coupling between spins and phonons is considered.

DOI: 10.1103/PhysRevB.75.052401 PACS number(s): 75.10.Jm, 75.10.Pq, 75.50.Ee

I. INTRODUCTION

As pointed out by Affleck,[1] it was quite a shock to the physics community when Haldane[2] argued in 1983 that there was a qualitative distinction between integral and half-integral spin antiferromagnetic Heisenberg chains. At first, the rather esoteric nature of Haldane's original arguments made the field rather impenetrable and, most of all, very attractive. Therefore, quantum *antiferromagnetism* became a very exciting field. For several reasons, it would be desirable to have quantum systems exhibiting a phase with Néel order separated from a phase with quantum disordered ground state by a critical fixed point. It is not always possible to find such systems but quantum spin *chains* represent a class of systems where it is natural to expect a quantum disordered ground state and, accordingly, a gap in the excitation spectrum. As shown by Haldane,[2] Heisenberg integer spin chains can be mapped into the quantum nonlinear σ model (NLσM) and the quantum critical fixed point occurs at zero coupling g^*. It is well known that the $O(3)$ σ model is a relativistic theory of a triplet of massive interacting particles. The gap corresponds, then, to the mass of the triplet which is proportional to the inverse of the correlation length, and, therefore, it is expected to increase with temperature.[3] The temperature dependence of the gap with temperature has been measured experimentally,[7,8,10,9] and the results agree qualitatively with the predictions obtained by using the NLσM.

By now, the existence of an excitation gap and a finite correlation length in integer spin one-dimensional Heisenberg antiferromagnets (1DHAF)—but not in half-integer spin chains—is very well established theoretically,[1] numerically,[4–6] and experimentally.[7–10] The numerical estimate for the gap energy for the 1DHAF, with spin 1, is $\Delta = 0.4105J$.

The peculiar behavior of 1DHAF contributed to enhancing the research on low-dimensional magnetism, and in the last two decades, we have witnessed remarkable discoveries of quantum phenomena in strongly correlated systems in low dimensions. It is now known that, besides the 1DHAF with integer spin—also called Haldane-gap system—there are other low-dimensional Heisenberg antiferromagnets (AFs) exhibiting a finite energy gap above the singlet ground state. Examples of such systems are the spin $S=1/2$ linear chain with bond alternation (dimerized chain or spin Peierls system[11]) and the spin $S=1/2$ two-leg spinladder. The 1DHAF system with integer spin can also have a gapless quantum phase when an external magnetic field H between the first H_{c1} and second critical field H_{c2} is applied. At $H = H_{c1}$, the system starts to develop magnetization and the gap is destroyed; at $H = H_{c2}$ the system is completely magnetized. Nowadays, there has been great interest in understanding the spin dynamics of this field-induced gapless quantum phase in 1DHAF.[12]

The experimental research in this field discovered materials that could not always fit exactly in the models studied theoretically. The $CsNiCl_3$ compound,[7] for example, is highly isotropic in its spin coupling but is only moderately one dimensional, while the compound known as $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ (NENP) is much more one dimensional but shows significant planar anisotropy.[8] As cited above, there is also great interest in understanding the behavior of low-dimensional magnets when external magnetic fields are applied. Therefore, a great number of theoretical studies have been devoted to the understanding of the properties of low-dimensional AFs when features such as interchain coupling, anisotropies, and external fields are included in the model. Particularly, the Haldane gap in spin $S=1$ HAF chains when terms describing an easy-axis anisotropy,[2] an applied magnetic field,[13,14] exchange and/or single-site anisotropies,[15] and dimerization[15] are included has been the subject of many works. However, the effect that the coupling between the spins of the magnetic system and the phonons of the lattice can have on the energy of the Haldane gap has not been taken into account. This is the aim of the present work.

However, we may expect that the spin-phonon coupling can influence the properties of one-dimensional HAF systems and, in particular, the energy gap. It is well known that an AF spin-1/2 chain coupled to optical phonons develops a spin gap via a static deformation or dimerization: this is known as the spin-Peierls instability.[16,17] For temperatures lower than T_{SP}, the spin-Peierls transition temperature, the system stays in the dimerized phase and for $T > T_{SP}$, the chain becomes undistorted and the gapless spin excitations are recovered. This transition has been observed in several quasi-one-dimensional organic materials[18] and its existence or not in the inorganic material $CuGeO_3$ has been the subject of a strong controversy. From the theoretical point of view, most of the treatments directed to spin-Peierls systems[11,16,17] have considered the phonons in a mean-field context expected to work when the phonon frequency can be neglected in comparison to the spin gap. Therefore, mean-field treatment is better suited to softer materials and cannot describe systems in which high-frequency phonons can couple to spin excitations.[19] Recently, Orignac and Chitra[20] proposed a method that is able to give a more quantitative description of the mean-field theory of the spin-Peierls transition, obtaining results for the thermodynamic properties of the system in a whole range of temperature.

Although Schulz[21] has shown that we cannot expect a spin-Peierls instability for integer spin chains, we have reason to expect that the effects due to the coupling between phonons and spin excitations can be rather important in low-dimensional magnetism. In this work, we will study the effect of this coupling on the energy gap of 1DHAF systems with spin 1. The behavior of these systems is mainly dominated by short-range fluctuations and the spin-phonon coupling is an important source of frustration in the quantum system. In addition, the study of such 1D systems must be considered as an important step for understanding the fluctuations intervening in two-dimensional systems (showing a vanishing critical temperature) because of the close connection with the high-T_c superconductors.

This Brief Report is organized as follows. The model Hamiltonian is presented in Sec. II, where we also discuss the approximations used in our treatment. The results for the gap energy dependence on the parameters related to the spin-phonon coupling are given in Sec. III. Our conclusions are presented in Sec. IV.

II. MODEL

We start from the following Hamiltonian for the spin-phonon coupling system on a linear chain,

$$\mathcal{H} = \sum_{lonA} \{[J + \alpha(u_l - u_{l+a})]\mathbf{S}_l \cdot \mathbf{S}_{l+a} + [J + \alpha(u_{l-a} - u_l)]\mathbf{S}_{l-a} \cdot \mathbf{S}_l\} + \mathcal{H}_{ph}, \tag{1}$$

where we assume that the lattice is divided in two sublattices A and B and the sum runs over all sites of lattice A. J is the usual exchange integral, $\mathbf{S}_l$ ($\mathbf{S}_{l\pm a}$) is the spin-1 operator on site l ($l\pm a$) of sublattice A (B), a is the lattice constant, α is the spin-phonon coupling, and u_l is the displacement along the chain. The harmonic lattice Hamiltonian is given by

$$\mathcal{H}_{ph} = \sum_{lonA} \left\{ \frac{M}{2}[(\dot{u}_l)^2 + (\dot{u}_{l+a})^2] + \frac{K}{2}[(u_l - u_{l+a})^2 + (u_{l-a} - u_l)^2] \right\}, \tag{2}$$

where M is the mass of the magnetic ion, K is the spring constant, and $\dot{u} = du/dt$.

This Hamiltonian, in two dimensions, was studied by Su and Zheng[22] by using the linearized spin-wave approximation in the two sublattice spaces and the second quantization representation for phonon operators. The Hamiltonian is then written as a sum of three terms,

$$\mathcal{H} = \mathcal{H}_{sw} + \mathcal{H}_{ph} + \mathcal{H}_1, \tag{3}$$

where $\mathcal{H}_{sw}$ and $\mathcal{H}_{ph}$ correspond, respectively, to the contributions from spin waves and from phonons up to second order and $\mathcal{H}_1$ corresponds to the interactions between these two excitations.

Writing the spin components in each sublattice in terms of spin-wave operators A_l and $B_{l\pm a}$,

$$S_l^{\dagger} = \sqrt{2S}A_l, \quad S_l^{-} = \sqrt{2S}A_l^{\dagger},$$

$$S_l^z = S - A_l^{\dagger}A_l, \quad S_{l+a}^z = -S + B_{l+a}^{\dagger}B_{l+a}, \tag{4}$$

$$S_{l+a}^{\dagger} = \sqrt{2S}B_{l+a}^{\dagger}, \quad S_{l+a}^{-} = \sqrt{2S}B_{l+a}, \tag{5}$$

we obtain

$$\mathcal{H}_{sw} = -NJ + 2J\sum_k \{A_k^{\dagger}A_k + B_k^{\dagger}B_k + \cos(k)[A_k^{\dagger}B_k^{\dagger} + B_kA_k]\}, \tag{6}$$

where N is the number of lattice sites, and A_k and B_k are the Fourier transforms of A_l and $B_{l\pm a}$. Following Su and Zheng,[22] we obtain for the phonon contribution,

$$\mathcal{H}_{ph} = \sum_q [\omega_c(q)c_q^{\dagger}c_q + \omega_d(q)d_q^{\dagger}d_q + 2], \tag{7}$$

where c_q and d_q are the phonon operators with frequencies

$$\omega_c(q) = \omega_D\left|\sin\left(\frac{q}{2}\right)\right|,$$

$$\omega_d(q) = \omega_D\left|\cos\left(\frac{q}{2}\right)\right|. \tag{8}$$

Here, we have used $\omega_D = \sqrt{2K/M}$.

For the interaction term, we write

$$\mathcal{H}_1 = -i\frac{\alpha}{\sqrt{MN}}\sum_{k,q}\left\{\frac{\sin(q/2)}{\sqrt{\omega_c(q)}}(c_{-q}^{\dagger} + c_q)\Gamma_{k,q} - \frac{\cos(q/2)}{\sqrt{\omega_d(q)}}(d_{-q}^{\dagger} + d_q)\Delta_{k,q}\right\}, \tag{9}$$

where we have defined[22]

$$\Gamma_{k,q} = \cos\left(\frac{q}{2}\right)(A_{k+q}^{\dagger}A_k + B_{k+q}B_k^{\dagger}) + \cos\left(k + \frac{q}{2}\right)(A_{k+q}^{\dagger}B_k^{\dagger} + B_{k+q}A_k),$$

$$\Delta_{k,q} = \sin\left(\frac{q}{2}\right)(A_{k+q}^{\dagger}A_k - B_{k+q}B_k^{\dagger}) + \sin\left(k + \frac{q}{2}\right)(A_{k+q}^{\dagger}B_k^{\dagger} - B_{k+q}A_k). \tag{10}$$

The next step is to use a Bogoliubov transformation for the spin-wave operators in order to make $\mathcal{H}_{sw}$ diagonal, that is,

$$A_k = \cosh(\theta_k)\alpha_k + \sinh(\theta_k)\beta_k^{\dagger},$$

$$B_k = \sinh(\theta_k)\alpha_k^{\dagger} + \cosh(\theta_k)\beta_k. \tag{11}$$

The expression for θ_k is determined in the context of the *modified spin-wave theory*[23] (MSWT). We obtain

$$\cosh(2\theta_k) = \frac{1}{\sqrt{1 - \eta^2\cos^2(k)}},$$

$$\sinh(2\theta_k) = -\frac{\eta\cos(k)}{\sqrt{1 - \eta^2\cos^2(k)}}. \tag{12}$$

The parameter η is determined by imposing the total staggered magnetization to be zero,[23] this is, basically, the essence of the MSWT. This restriction leads to

$$\sum_{l} (A_l^\dagger A_l + B_{l+a}^\dagger B_{l+a}) = NS. \tag{13}$$

Finally, we obtain for the spin-wave Hamiltonian the expression

$$\mathcal{H}_{sw} = \frac{2JS}{\eta}\sum_{k}(\epsilon_k - 1) + \frac{2JS}{\eta}\sum_{k}\epsilon_k(\alpha_k^\dagger\alpha_k + \beta_k^\dagger\beta_k), \tag{14}$$

where

$$\epsilon_k = \sqrt{1-\eta^2\cos^2(k)}. \tag{15}$$

Solving Eq. (13) for the Hamiltonian [Eq. (3)] leads to the following equation:[22]

$$\frac{1}{2N}\sum_{k}\xi_k\Bigg(1+\frac{\alpha^2}{NK\omega_D}\sum_{q}(\zeta_k^c\{1+\cos(2k+q)-\eta\cos(k)\times[\cos(k)+\cos(k+q)]\}+\zeta_k^d\{1-\cos(2k+q)+\eta\cos(k)\times[\cos(k+q)-\cos(k)]\})\Bigg)=1, \tag{16}$$

where the definitions

$$\xi_k = \frac{\coth[J\sqrt{1-\eta^2\cos^2(k)}/(T\eta)]}{\sqrt{1-\eta^2\cos^2(k)}},$$

$$\zeta_k^c = \frac{|\sin(q/2)|}{\{|\sin(q/2)|+[2J/(\eta\omega_D)][\sqrt{1-\eta^2\cos^2(k)}+\sqrt{1-\eta^2\cos^2(k+q)}]\}^2},$$

$$\zeta_k^d = \frac{|\cos(q/2)|}{\{|\cos(q/2)|+[2J/(\eta\omega_D)][\sqrt{1-\eta^2\cos^2(k)}+\sqrt{1-\eta^2\cos^2(k+q)}]\}^2} \tag{17}$$

were used.[24] It can be seen that the resulting Hamiltonian is temperature dependent because the parameter ζ depends on T: the effect of the temperature is to renormalize the parameters of the system when we consider interactions. The MSWT is, in a sense, a self-consistent harmonic approximation where we substitute the exact Hamiltonian by an harmonic one with renormalized, temperature-dependent, parameters. Thus, our estimate for the temperature dependence of the gap is to be understood in the same spirit as the one calculated by Jolicoeur and Golinelli.[3]

In order to obtain the spin-wave energy, our task in this work, we numerically solved the self-consistent equation [Eq. (15)] to obtain the parameter η. The expression for the spin-wave energy is then

$$\varepsilon(k) = \frac{JS}{\eta}\sqrt{1-\eta^2\cos^2(k)}. \tag{18}$$

The energy gap corresponds to $\varepsilon(k=0)=\Delta$ and depends on the temperature T, as explained above, on the spin-phonon coupling parameter α and on the frequency ω_D, which is determined by microscopic properties of the lattice as the spring constant K and the mass M of the magnetic ion.

III. RESULTS

If we neglect the spin-phonon interaction, that is, taking $\alpha=0$, we obtain the gap energy for the 1DHAF studied by Haldane.[2] For $T=0$ and $S=1$, the value estimated by the MSW theory is $\Delta(T=0)=\Delta_0=0.178J$, which is in close agreement with the value of 0.17 obtained by Arovas and Auerbach[25] by using the Schwinger boson method. For $\alpha=0$ and $T=0$, Eq. (16) becomes

$$S = \frac{1}{\pi}K(\eta), \tag{19}$$

where $K(\eta)$ is the complete elliptic integral of the first kind. Using asymptotic expansions for $K(\eta)$, we obtain

$$\Delta_0 \approx 8Se^{-\pi S}, \tag{20}$$

for the gap dependence on the spin value at $T=0$. This behavior is in good agreement with the result obtained by applying the renormalization group technique on the NLσ model, which gives $\Delta_0=CS^2\exp(-\pi S)$, where C is a constant independent of the spin value. This remarkable agreement between two completely different theories, the MSW and the renormalization group, was pointed out by Arovas and Auerbach.[25] However, it is important to mention, as discussed in Ref. 25, that the MSW approach has some drawbacks, for example, the inability to explain the gaplessness of all half-odd-integer AFs chains.

Despite the fact that the MSW and Schwinger boson method estimates for the gap energy agree quite well, we have to note that this estimated value of $\approx 0.18J$ is quite smaller than the numerical value of $0.41J$. As it is very well known, up to the present moment, *no* analytical approach, including the NLσ model and more sophisticated theories, is

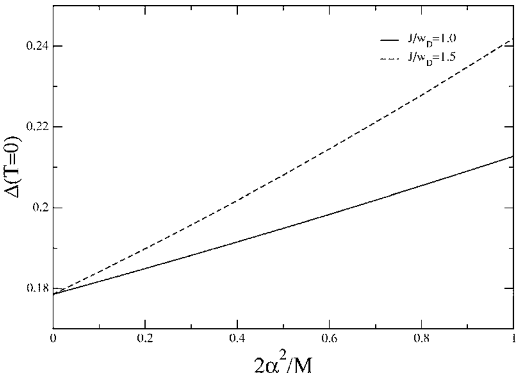

FIG. 1. Gap at zero temperature, $\Delta(T=0)$, as a function of the spin-phonon coupling α. The results for two different values of the parameter ω_D are shown.

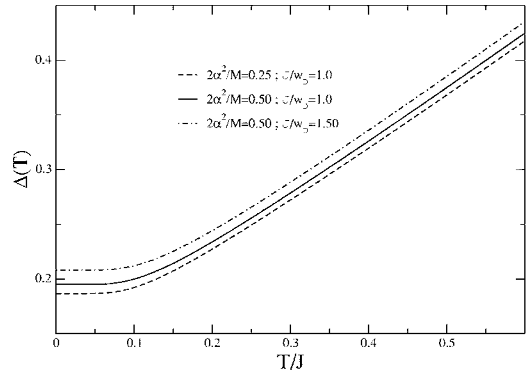

FIG. 2. Gap, $\Delta(T)$, as a function of the temperature: the continuous curve corresponds to $\alpha=0.5$ and $J\omega_D=1.0$, the dashed line is for $\alpha=0.25$ and $J\omega_D=1.0$, and the dot-dashed line corresponds to $\alpha=0.50$ and $J\omega_D=1.5$.

able to predict the correct value for Δ_0. Therefore, what is usually obtained in theoretical approaches is a set of expressions for finite temperatures involving ratios such as $\Delta(T)/\Delta_0$ and T/Δ_0.

We now turn back to the main subject of this work: the influence of the spin-phonon coupling on the gap energy. In Eq. (16), we see that α, the parameter describing the strength of the spin-phonon coupling, appears combined with K and ω_D. In order to isolate the effects due to α, the spin-coupling parameter, and the elastic properties of the chain (assuming M as a constant), we write $\alpha^2/(K\omega_D)=2\alpha^2/(M\omega_D^3)$ and perform our calculations varying $2\alpha^2/M$ and J/ω_D. Figure 1 shows the gap energy at $T=0$ as a function of $2\alpha^2/M$ for two different values of ω_D. First, we see that the effect of the spin-phonon coupling is to stabilize the gap because the energy increases with α. The coupling influence is greater for smaller ω_D, as could be expected: a large value of ω_D means a more rigid system, in which the interaction between spin waves and the phonons of the system is low: in fact, $\omega_d \to \infty$ must recover the incompressible $(\alpha=0)$ chain results.

The temperature dependence of the gap, for three pairs of α and ω_D values, is shown in Fig. 2, where it can be seen that the temperature dependence of the gap does not vary appreciably as these parameters are changed. The dashed and continuous curves correspond to the same spring constant (M taken as a constant) and to different values for α: as expected, the gap is larger for the largest value of the spin-coupling parameter. Again, comparing the dot-dashed ($\alpha=0.50$, $\omega_D=1.5$) and the continuous ($\alpha=0.50$, $\omega_D=1.0$) curves, we see that a softer chain provides a larger gap.

IV. CONCLUSIONS

In this work, we have considered the effect of the coupling between the spin waves of the 1DHAF model and the lattice phonons on the energy gap expected for the model. Our results show that the effect of this coupling is to enhance the gap. However, the temperature dependence of this gap is not affected by the coupling. We hope our results can stimulate further numerical investigation that can check these predictions.

ACKNOWLEDGMENT

This work was partially supported by CNPq (A.S.T.P. and M.E.G.).

*Electronic address: meg@dppg.cefetmg.br
[1] I. Affleck, J. Phys.: Condens. Matter **1**, 3047 (1989).
[2] F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983); Phys. Rev. Lett. **50**, 1153 (1983).
[3] Th. Jolicoeur and O. Golinelli, Phys. Rev. B **50**, 9265 (1994).
[4] I. Affleck *et al.*, Phys. Rev. Lett. **59**, 799 (1987).
[5] M. Takahashi, Phys. Rev. Lett. **62**, 2313 (1989).
[6] H. J. Schulz and T. Ziman, Phys. Rev. B **33**, 6545 (1986).
[7] W. J. L. Buyers *et al.*, Phys. Rev. Lett. **56**, 371 (1986).
[8] J. P. Renard *et al.*, Europhys. Lett. **3**, 945 (1987).
[9] T. Sakaguchi *et al.*, J. Phys. Soc. Jpn. **65**, 3025 (1996).
[10] A. Zheludev *et al.*, Phys. Rev. B **53**, 15004 (1996).
[11] M. C. Cross and D. S. Fisher, Phys. Rev. B **19**, 402 (1979).
[12] T. Goto *et al.*, Phys. Rev. B **73**, 214406 (2006).
[13] I. Affleck, Phys. Rev. B **41**, 6697 (1990).
[14] S. Sachdev *et al.*, Phys. Rev. B **50**, 258 (1994).
[15] Q. Jiang and X. Y. Chen, Phys. Lett. A **291**, 417 (2001).
[16] P. Pincus, Solid State Commun. **4**, 1971 (1971).
[17] E. Pytte, Phys. Rev. B **10**, 4637 (1974).
[18] The spin-Peierls transition has been observed in several quasi-1D organic materials: I. S. Jacobs *et al.*, Phys. Rev. B **14**, 3036 (1976); S. Huizinga *et al.*, *ibid.* **19**, 4723 (1979); A. Maaroufi *et al.*, Mol. Cryst. Liq. Cryst. **119**, 311 (1985); J. P. Pouget *et al.*, *ibid.* **79**, 129 (1982); R. Laversanne *et al.*, J. Phys. (France) Lett. **45**, L393 (1984); Q. Liu *et al.*, Synth. Met. **56**, 1840 (1993).
[19] C. Gros and R. Werner, Phys. Rev. B **58**, R14677 (1998).
[20] E. Orignac and R. Chitra, Phys. Rev. B **70**, 214436 (2004).
[21] T. Jolicoeur and O. Golinelli, Phys. Rev. B **50**, 9265 (1994).
[22] X. Su and H. Zheng, Solid State Commun. **109**, 323 (1999).
[23] S. Tang *et al.*, Phys. Rev. B **40**, 5000 (1989).
[24] H. Zheng, Phys. Lett. A **199**, 409 (1995).
[25] D. P. Arovas and A. Auerbach, Phys. Rev. B **38**, 316 (1988).