Fermionic versus bosonic descriptions of one-dimensional spin-gapped antiferromagnets

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In terms of spinless fermions and spin waves, we describe magnetic properties of a spin-$1/2$ ferromagnetic−antiferromagnetic bond-alternating chain which behaves as a Haldane-gap antiferromagnet. On one hand, we employ the Jordan−Wigner transformation and treat the fermionic Hamiltonian within the Hartree−Fock approximation. On the other hand, we employ the Holstein−Primakoff transformation and modify the conventional spin-wave theory so as to restore the sublattice symmetry. We calculate the excitation gap, the specific heat, the magnetic susceptibility, magnetization curves, and the nuclear spin-lattice relaxation rate with varying bond alternation. These schemes are further applied to a bond-alternating tetramerized chain which behaves as a ferrimagnet. The fermionic language is particularly stressed as a useful tool to investigate one-dimensional spin-gapped antiferromagnets, while the bosonic one works better for ferrimagnets.

1. Introduction

Haldane [1,2] sparked renewed interest in one-dimensional Heisenberg antiferromagnets, predicting that their low-energy structures should qualitatively vary according as the constituent spins are integral or fractional. A magnetic excitation gap immediately above the ground state, which is referred to as the Haldane gap, was indeed observed in quasi-one-dimensional spin-1 Heisenberg antiferromagnets such as $CsNiCl_3$ [3] and $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ [4,5]. A rigorous example of such a massive phase was also given theoretically [6,7]. Significant numerical efforts [8−12] were dovoted to detecting the Haldane gap in the higher-spin systems. Competition between massive and massless phases in low-dimensional quantum magnets was extensively studied especially by the nonlinear-sigma -model quantum field theory [13−23] and a wide variety of spin gaps-energy gaps in magnetic excitation spectra-were further predicted. There followed stimulative findings, including quantized plateaux in zero-temperature magnetization curves [24−26], gap formation in coupled spin chains [27,28] and the dramatic crossover from one- to two-dimensional quantum antiferromagnets [29], and an antiferromagnetic excitation gap with a ferromagnetic background [30−33].

Besides the sigma-model study, analytic approaches played a crucial role in revealing the nature of Haldane-gap antiferromagnets. The valence-bond-solid model [13,14] stimulated considerable interest in matrix-product representation [34−40] of the Haldane phase. The Lieb−Schultz−Mattis theorem [41] was generalized [24] to clarify a mechanism for gap formation in a magnetic field. However, these arguments were essentially restricted to the ground-state behavior and can hardly be extended to finite-temperature properties. Numerical tools such as quantum Monte Carlo and density-matrix renormalization-group techniques are indeed useful for such a purpose, but an analytic strategy is still indispensable to low-temperature thermodynamics especially of spin-gapped antiferromagnets, where grand canonical sampling is hardly feasible numerically. Then we are led to describe massive spin chains in terms of conventional languages such as the Jordan−Wigner fermions and the Holstein−Primakoff spin waves.

The Jordan−Wigner transformation is an efficient approach to low-dimensional quantum magnetism. Spin-$1/2$ arrays with uniform [42] and alternating [43−45] antiferromagnetic exchange interactions between nearest neighbors were thus investigated and their energy structures, magnetization curves, and

thermodynamic properties were indeed revealed well. Two-leg antiferromagnetic spin ladders were also discussed within this scheme [46,47] and the inter-chain-coupling effect on the lowest-lying excitation was elucidated. More refined fermionization was further proposed for coupled spin chains. Ordering spinless fermions along a snake-like path, Dai and Su [48] succeeded in interpreting massive and massless excitations with varying number of the ladder legs. Their idea was generalized to investigate zero-temperature magnetization curves [49] and thermodynamic quantities. [50] In such circumstances, we consider fermionizing an spin-1/2 ferromagnetic−antiferromagnetic bond-alternating chain, which converges to the spin-1 antiferromagnetic Heisenberg chain as the ferromagnetic coupling tends to infinity and therefore reproduces many of observations common to Haldane-gap antiferromagnets.

Bosonic theory has significantly been developed for one-dimensional quantum magnets in recent years. While the Schwinger-boson mean-field theory is unable to distinguish fractional-spin chains from integral-spin ones, it is still useful in predicting the asymptotic dependence of the Haldane gap on spin quantum number [51] and explaining quantum phase transitions of Haldane-gap antiferromagnets in a field [52]. The Schwinger-boson representation was further applied to ferrimagnetic spin chains [53,54] and ladders [55]. It was a major breakthrough leading to the subsequent development of the spin-wave theory in low dimensions [56−62] that Takahashi [63] gave a spin-wave description of the one-dimensional ferromagnetic thermodynamics introducing an additional constraint on the number of spin waves. This modified spin-wave scheme was further applied to spin-gapped antiferromagnets [50,64,65] and qualitatively improved for one-dimensional ferrimagnets [66,67]. The antiferromagnetic modified spin-wave theory is less quantitative than the ferrimagnetic version [54,68], but it enlighteningly interpreted novel observations such as the temperature dependence of the Haldane gap [64,65] and the field dependence of the nuclear spin-lattice relaxation rate [69]. Such spin-wave understanding is well supported by other analytic descriptions [70−72]. As for finite-temperature calculation of spin-gapped antiferromagnets, the Schwinger-boson mean-field theory is of no use, while the modified spin-wave theory maintains its validity to a certain extent [54]. Thus, we apply the modified spin-wave scheme to the spin-1/2 ferromagnetic−antiferromagnetic bond-alternating chain with particular emphasis on a comparison between fermionic and bosonic descriptions of spin-gapped antiferromagnets.

Our theoretical attempt is much motivated by existent bond-alternating chain compounds such as IPACuCl$_3$ (IPA = isopropylammonium = (CH$_3$)$_2$CHNH$_3$) [73] and (4-BzpipdH)CuCl$_3$ (4-BzpipdH = 4-benzylpiperidinium = C$_{12}$H$_{18}$N) [74]. These materials behave as spin-1 Haldane-gap antiferromagnets at low temperatures [75−78], while such spin-1 features are broken up into paramagnetic spin 1/2's with increasing temperature [79−81]. Besides the thermal crossover from quantum spin 1's to classical spin 1/2's, their enriched ground-state properties [82−86] and novel edge states [87] are of great interest to both theoreticians and experimentalists.

2. Formalism

We consider the ferromagnetic−antiferromagnetic bond-dimeric spin-1/2 Heisenberg chain, whose Hamiltonian is given by

$$\mathcal{H} = \sum_{n=1}^{N}[(J_{AF}\mathbf{S}_{2n-1}\cdot\mathbf{S}_{2n} - J_F\mathbf{S}_{2n}\cdot\mathbf{S}_{2n+1}) - $$
$$-g\mu_B H(S_{2n-1}^z + S_{2n}^z)]. \tag{1}$$

The ground-state properties [88−91] and low-lying excitations [92] of this model were well investigated by numerical tools and variational schemes. In particular, the string order parameter originally defined for spin-1 Heisenberg chains [93] was generalized to this system [88−90] and the breakdown of a hidden $Z_2 \times Z_2$ symmetry was extensively argued [84,85]. As the ferromagnetic coupling tends to infinity, the string order remains finite and the Haldane gap converges to that originating in decoupled singlet dimers.

On the other hand, the thermodynamic properties have much less been calculated so far [87,94] and there is no guiding theory for extensive experimental findings. Employing two different languages, we calculate various thermal quantities and give rigorous information on their low-temperature behavior.

1.1. Fermionic approach

In accordance with the bond dimerization, we introduce two kinds of spinless fermions through the Jordan−Wigner transformation

$$S_{2n-1}^+ = a_n^\dagger \exp\left[i\pi\left(\sum_{m=1}^{n-1}a_m^\dagger a_m + \sum_{m=1}^{n-1}b_m^\dagger b_m\right)\right],$$

$$S_{2n}^+ = b_n^\dagger \exp\left[i\pi\left(\sum_{m=1}^{n}a_m^\dagger a_m + \sum_{m=1}^{n-1}b_m^\dagger b_m\right)\right],$$

$$S_{2n-1}^z = a_n^\dagger a_n - \frac{1}{2}, \quad S_{2n}^z = b_n^\dagger b_n - \frac{1}{2}. \tag{2}$$

Decomposing the fermionic Hamiltonian at the Hartree−Fock level, we obtain a mean-field Hamiltonian as

$$\mathcal{H}_{HF} = E_0 + (J_{AF} - J_F) \times$$

$$\times \sum_{n=1}^{N} \left[\left(d_b - \frac{1}{2} \right) a_n^\dagger a_n + \left(d_a - \frac{1}{2} \right) b_n^\dagger b_n \right] +$$

$$+ J_{AF} \sum_{n=1}^{N} \left[\left(\frac{1}{2} - p_{AF} \right) a_n^\dagger b_n + \text{H.c.} \right] -$$

$$- J_F \sum_{n=1}^{N} \left[\left(\frac{1}{2} - p_F \right) b_n^\dagger a_{n+1} + \text{H.c.} \right] -$$

$$- g\mu_B H \sum_{n=1}^{N} (a_n^\dagger a_n + b_n^\dagger b_n), \qquad (3)$$

where $d_a = \langle a_n^\dagger a_n \rangle_{HF}$, $d_b = \langle b_n^\dagger b_n \rangle_{HF}$, $p_{AF} = \langle b_n^\dagger a_n \rangle_{HF}$, $p_F = \langle a_{n+1}^\dagger b_n \rangle_{HF}$, and

$$E_0 = \left[J_{AF} \left(|p_{AF}|^2 - d_a d_b + \frac{1}{4} \right) - \right.$$

$$\left. - J_F \left(|p_F|^2 - d_a d_b + \frac{1}{4} \right) + g\mu_B H \right] N, \qquad (4)$$

with $\langle \ldots \rangle_{HF}$ denoting the thermal average over the Hartree−Fock eigenstates. Defining the Fourier transformation as

$$a_n = \frac{1}{\sqrt{N}} \sum_k e^{ik(n-1/4)} a_k,$$

$$b_n = \frac{1}{\sqrt{N}} \sum_k e^{ik(n+1/4)} b_k \qquad (5)$$

and then a unitary transformation as

$$\begin{pmatrix} a_k \\ b_k \end{pmatrix} = \begin{pmatrix} u_k & v_k e^{i\theta_k} \\ v_k e^{-i\theta_k} & -u_k \end{pmatrix} \begin{pmatrix} \alpha_k \\ \beta_k \end{pmatrix}, \qquad (6)$$

where

$$u_k = \sqrt{\frac{1}{2} \left(1 - \frac{\eta}{\sqrt{\eta^2 + |\gamma_k|^2}} \right)}, \ v_k = \sqrt{\frac{1}{2} \left(1 + \frac{\eta}{\sqrt{\eta^2 + |\gamma_k|^2}} \right)},$$

$$\gamma_k \equiv |\gamma_k| e^{i\theta_k} =$$

$$= J_{AF} \left(\frac{1}{2} - p_{AF} \right) e^{ik/2} - J_F \left(\frac{1}{2} - p_F^* \right) e^{-ik/2},$$

$$\xi = \frac{1}{2} (J_{AF} - J_F)(d_a + d_b - 1),$$

$$\eta = \frac{1}{2} (J_{AF} - J_F)(d_a - d_b), \qquad (7)$$

and twice the lattice constant is set equal to unity, we can diagonalize the Hamiltonian as

$$\mathcal{H}_{HF} = E_0 + \sum_k (\varepsilon_k^+ \alpha_k^\dagger \alpha_k + \varepsilon_k^- \beta_k^\dagger \beta_k), \qquad (8)$$

where the dispersion relations are given by

$$\varepsilon_k^\pm = \xi \pm \sqrt{\eta^2 + |\gamma_k|^2} - g\mu_B H. \qquad (9)$$

In terms of the fermion distribution functions $\bar{n}_k^\pm = [e^{\varepsilon_k^\pm / k_B T} + 1]^{-1}$, the internal energy, the total magnetization, and the magnetic susceptibility are expressed as

$$E = E_0 + \sum_k \sum_{\sigma = \pm} \varepsilon_k^\sigma \bar{n}_k^\sigma, \qquad (10)$$

$$M = \sum_k \sum_{\sigma = \pm} \bar{n}_k^\sigma - N, \qquad (11)$$

$$\chi = \frac{(g\mu_B)^2}{k_B T} \sum_k \sum_{\sigma = \pm} \bar{n}_k^\sigma (1 - \bar{n}_k^\sigma), \qquad (12)$$

respectively. Another quantity of wide interest is the nuclear spin-lattice relaxation rate $1/T_1$. Considering the electronic-nuclear energy-conservation requirement, the Raman process usually plays a leading role in the relaxation, which is formulated as

$$\frac{1}{T_1} = \frac{4\pi (g\mu_B \hbar \gamma_N)^2}{\hbar \sum_m e^{-E_m/k_B T}} \sum_{m,m'} e^{-E_m/k_B T} \times$$

$$\times \left| \langle m' | \sum_n (A_n S_{2n-1}^z + B_n S_{2n}^z) | m \rangle \right|^2 \delta(E_{m'} - E_m - \hbar\omega_N),$$

$$(13)$$

where A_n and B_n are the dipolar coupling constants between the nuclear and electronic spins, $\omega_N = \gamma_N H$ is the Larmor frequency of the nuclei with γ_N being the gyromagnetic ratio, and the summation $\sum_m$ is taken over all the electronic eigenstates $|m\rangle$ with energy E_m. Assuming the Fourier components of the coupling constants to have little momentum dependence as $\sum_n \exp(ikn) A_n \equiv A_k \simeq A$ and $\sum_n \exp(ikn) B_n \equiv B_k \simeq B$, we obtain the fermionic expression of the Raman relaxation rate as

$$\frac{1}{T_1} = \pi (g\mu_B \hbar \gamma_N)^2 / \hbar N^2 \times$$

$$\times \sum_{k,k'} [A^2 + B^2 + 2AB \cos(\theta_{k'} - \theta_k)] \times$$

$$\times \sum_{\sigma = \pm} \bar{n}_k^\sigma (1 - \bar{n}_{k'}^\sigma) \delta(\varepsilon_{k'}^\sigma - \varepsilon_k^\sigma - \hbar\omega_N). \qquad (14)$$

1.2. Bosonic approach

Next we consider a single-component bosonic representation of each spin variable at the cost of the rota-

tional symmetry. We start from the Holstein−Primakoff transformation

$$S^+_{4n-4+\tau} = \sqrt{2S - a^\dagger_{\tau:n} a_{\tau:n}}\, a_{\tau:n},$$

$$S^z_{4n-4+\tau} = S - a^\dagger_{\tau:n} a_{\tau:n},$$

$$S^+_{4n-2+\tau} = b^\dagger_{\tau:n} \sqrt{2S - b^\dagger_{\tau:n} b_{\tau:n}},$$

$$S^z_{4n-2+\tau} = -S + b^\dagger_{\tau:n} b_{\tau:n}, \tag{15}$$

where $\tau = 1,2$; that is, we assume the chain to consist of four sublattices. Under the large-S treatment, the Hamiltonian can be expanded as

$$\mathcal{H} = -2(J_F + J_{AF})S^2 N + E_1 + E_0 +$$
$$+ \mathcal{H}_1 + \mathcal{H}_0 + O(S^{-1}), \tag{16}$$

where E_i and H_i give the $O(S^i)$ quantum corrections to the ground-state energy and the dispersion relations, respectively. The naivest diagonalization of the Hamiltonian (16), whether up to $O(S^1)$ or up to $O(S^0)$, results in diverging sublattice magnetizations even at zero temperature. In order to suppress the quantum as well as thermal divergence of the number of bosons, we consider minimizing the free energy constraining the sublattice magnetizations to be zero [56−58]:

$$\sum_{n=1}^{N} \sum_{\tau=1,2} (a^\dagger_{\tau:n} a_{\tau:n} + b^\dagger_{\tau:n} b_{\tau:n}) = 4SN. \tag{17}$$

Within the conventional spin-wave theory, spins on one sublattice point predominantly up, while those on the other predominantly down. The condition (17) restores the sublattice symmetry. In order to enforce the constraint (17), we first introduce a Lagrange multiplier and diagonalize

$$\mathcal{H} = \mathcal{H} + J_{AF}\nu S \sum_{n=1}^{N} \sum_{\tau=1,2} (a^\dagger_{\tau:n} a_{\tau:n} + b^\dagger_{\tau:n} b_{\tau:n}). \tag{18}$$

We define the Fourier transformation as

$$a_{\tau:n} = \frac{1}{\sqrt{N}} \sum_k e^{-ik(n-5/8+\tau/4)} a_{\tau:k},$$

$$b_{\tau:n} = \frac{1}{\sqrt{N}} \sum_k e^{ik(n-1/8+\tau/4)} b_{\tau:k}, \tag{19}$$

and then the Bogoliubov transformation as

$$\begin{pmatrix} a_{1:k} \\ a_{2:k} \\ b^\dagger_{1:k} \\ b^\dagger_{2:k} \end{pmatrix} = \begin{pmatrix} \psi^+_{1:k} & \psi^-_{1:k} & -\psi^{+*}_{4:k} & -\psi^{-*}_{4:k} \\ \psi^+_{2:k} & \psi^-_{2:k} & -\psi^{+*}_{3:k} & -\psi^{-*}_{3:k} \\ -\psi^+_{3:k} & -\psi^-_{3:k} & \psi^{+*}_{2:k} & \psi^{-*}_{2:k} \\ -\psi^+_{4:k} & -\psi^-_{4:k} & \psi^{+*}_{1:k} & \psi^{-*}_{1:k} \end{pmatrix} \begin{pmatrix} \alpha_{1:k} \\ \beta_{1:k} \\ \alpha^\dagger_{2:k} \\ \beta^\dagger_{2:k} \end{pmatrix}, \tag{20}$$

where four times the lattice constant is set equal to unity. We determine the coefficients $\psi^\pm_{i:k}$ so as to diagonalize $\mathcal{H}$ up to the order of $O(S)$ and perturbationally take $\mathcal{H}_0$ into calculation [95]. Then the Hamiltonian (16) is written as

$$E_1 = -2J_{AF}(1 + \gamma + \nu)SN + J_{AF} \sum_k \sum_{\sigma=\pm} \omega^\sigma_k,$$

$$E_0 = 2J_{AF}[2\Delta(\Lambda - \gamma\Gamma) - (1+\gamma)\Delta^2 - \gamma\Gamma^2 - \Lambda^2],$$

$$\mathcal{H}_1 = J_{AF} \sum_k \sum_{\tau=1,2} (\omega^+_k \alpha^\dagger_{\tau:k} \alpha_{\tau:k} + \omega^-_k \beta^\dagger_{\tau:k} \beta_{\tau:k}),$$

$$\mathcal{H}_0 = J_{AF} \sum_k \sum_{\tau=1,2} (\delta\omega^+_k \alpha^\dagger_{\tau:k} \alpha_{\tau:k} + \delta\omega^-_k \beta^\dagger_{\tau:k} \beta_{\tau:k}) +$$
$$+ \mathcal{H}_{\text{irrel}} + \mathcal{H}_{\text{quart}}, \tag{21}$$

where $\gamma = J_F/J_{AF}$,

$$\omega^\sigma_k = S\sqrt{(1+\gamma+\nu)^2 - 1 + \gamma^2 + 2\sigma\chi_k},$$

$$\delta\omega^\sigma_k = [(\Lambda - \gamma\Gamma) - (1+\gamma)\Delta]\left(1 + \frac{\sigma\gamma}{\chi_k}\right)\frac{1+\gamma+\nu}{\lambda^\sigma_k} -$$

$$- \gamma(\Delta + \Gamma)\frac{\gamma + \sigma\chi_k}{\lambda^\sigma_k} + (\Delta - \Lambda)\frac{\sigma\chi_k + \gamma\sin^2\frac{k}{2}}{\sigma\chi_k\lambda^\sigma_k},$$

$$\Gamma = \frac{1}{4N} \sum_k \sum_{\sigma=\pm} \left[\left(1 + \frac{\sigma\gamma}{\chi_k}\right)\frac{1+\gamma+\nu}{\lambda^\sigma_k} - 1\right] \times$$

$$\times \frac{(1+\gamma+\nu)\sigma\chi_k\cos^2\frac{k}{2} - \gamma(\gamma + \sigma\chi_k)\lambda^\sigma_k\sin^2\frac{k}{2}}{\chi^2_k + \gamma(\gamma + 2\sigma\chi_k)\sin^2\frac{k}{2}},$$

$$\Lambda = \frac{1}{4N} \sum_k \sum_{\sigma=\pm} \frac{\sigma\chi_k + \gamma\sin^2\frac{k}{2}}{\sigma\chi_k\lambda^\sigma_k},$$

$$\Delta = \frac{1}{4N} \sum_k \sum_{\sigma=\pm} \left[\left(1 + \frac{\sigma\gamma}{\chi_k}\right)\frac{1+\gamma+\nu}{\lambda^\sigma_k} - 1\right], \tag{22}$$

with $\lambda^\sigma_k = \omega^\sigma_k/S$

and $\chi_k = [(1+\gamma+\nu)^2 - \sin^2(k/2)]^{1/2}$.
$\mathcal{H}_{\text{irrel}}$ and $\mathcal{H}_{\text{quart}}$ in $\mathcal{H}_0$ contain off-diagonal one-body terms such as $\alpha_{\tau:k}\beta_{\tau:k}$ and residual

two-body interactions, respectively, both of which are neglected in the perturbational treatment.

At finite temperatures we replace $\alpha_{\tau:k}^\dagger \alpha_{\tau:k}$ and $\beta_{\tau:k}^\dagger \beta_{\tau:k}$ by their canonical averages $\langle \alpha_{\tau:k}^\dagger \alpha_{\tau:k} \rangle \equiv \bar{n}_{\tau:k}^+$ and $\langle \beta_{\tau:k}^\dagger \beta_{\tau:k} \rangle \equiv \bar{n}_{\tau:k}^-$, respectively, which are expressed as $\bar{n}_{\tau:k}^\pm \equiv \bar{n}_k^\pm = [e^{J_{AF}(\omega_k^\sigma + \delta\omega_k^\sigma)/k_B T} - 1]^{-1}$. Here the Lagrange multiplier ν is determined through

$$\sum_k \sum_{\sigma=\pm} \left(1 + \frac{\sigma\gamma}{\chi_k}\right)\frac{1+\gamma+\nu}{\lambda_k^\sigma}(1+2\bar{n}_k^\sigma) = 2N(1+2S). \tag{23}$$

Then the internal energy and the magnetic susceptibility are given by [96]

$$E = E_g + 2\sum_k \sum_{\sigma=\pm} \tilde{\omega}_k^\sigma \bar{n}_k^\sigma,$$

$$\chi = \frac{2(g\mu_B)^2}{3k_B T}\sum_k \sum_{\sigma=\pm} \bar{n}_k^\sigma(\bar{n}_k^\sigma + 1), \tag{24}$$

where $E_g = -2(J_F + J_{AF})S^2 N + E_1 + E_0$.

3. Calculations

First we calculate the ground-state energy E_g and the lowest excitation gap E_{gap} and compare them with numerical findings in Fig. 1. The spinless fermions are much better than the modified spin waves at describing both quantities. As J_F goes to zero, the fermionic findings are refined and end up with the exact values $E_g/N = -3J_{AF}/4$ and $E_{gap} = J_{AF}$. The modified spin waves considerably underestimate the spin gap. They can not distinguish massive spin chains from massless critical ones [54] to begin with, but they are still useful in qualitatively investigating dependences of the Haldane gap on temperature and spin quantum number [65].
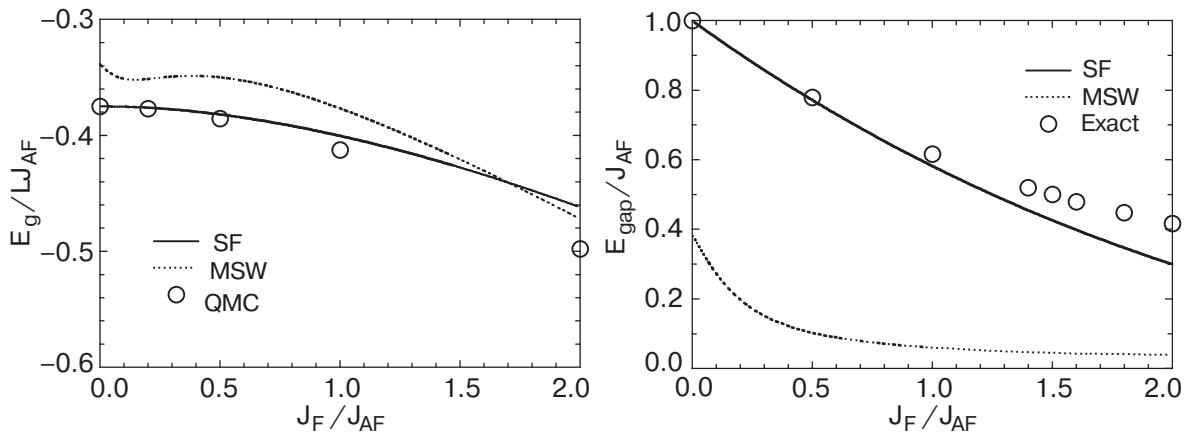
Secondly we calculate the thermodynamic properties. Figure 2 shows the temperature dependences of the zero-field specific heat and magnetic susceptibility. Due to the significant underestimate of the spin gap, the modified spin-wave description is much less quantitative than the fermionic one at low temperatures. Furthermore, the modified spin waves completely fail to reproduce the antiferromagnetic Schottky-type peak of the specific heat. Because of the Lagrange multiplier ν, which turns out a monotonically increasing function of temperature, the dispersion relations (22) lead to endlessly increasing energy and thus nonvanishing specific heat at high temperatures. The spinless fermions succeed in reproducing the overall thermal behavior. The present approaches have the advantage of giving the low-temperature behavior analytically. Equation (9) shows that the dispersion relation of the low-lying excitations reads

$$\varepsilon_k^\pm \simeq \pm(E_{gap} + J_{AF}\upsilon k^2) - g\mu_B H, \tag{25}$$

provided $g\mu_B H < E_{gap}$, where

$$E_{gap} = \sqrt{J_{AF}^2 \tilde{p}_{AF}^2 + J_F^2 \tilde{p}_F^2 + 2J_F J_{AF} \tilde{p}_F \tilde{p}_{AF}},$$

$$2E_{gap}\upsilon = J_F \tilde{p}_F \tilde{p}_{AF}, \tag{26}$$

with $\tilde{p}_F = \text{Re}\, p_F - 1/2$ and $\tilde{p}_{AF} = \text{Re}\, p_{AF} - 1/2$. Then the low-temperature properties are calculated as

$$\frac{C}{Nk_B} \simeq \sqrt{\frac{k_B T}{\pi \upsilon J_{AF}}} e^{-E_{gap}/k_B T}\left[\left(\frac{E_{gap}}{k_B T}\right)^2 + \frac{E_{gap}}{k_B T} + \frac{3}{4}\right],$$

$$\frac{\chi J_{AF}}{(g\mu_B)^2 N} \simeq \sqrt{\frac{J_{AF}}{\pi \upsilon k_B T}} e^{-E_{gap}/k_B T}. \tag{27}$$

These features are found in the antiferromagnetic Heisenberg two-leg ladder as well [50,72] and can be regarded as common to spin-gapped antiferromagnets.
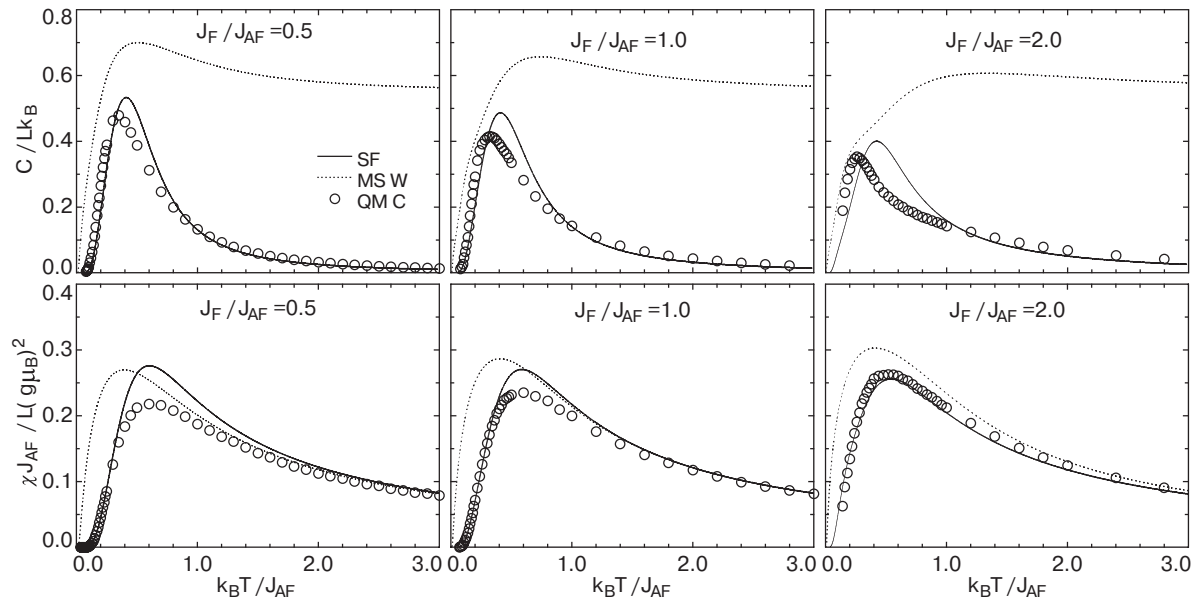

Fig. 1. The spinless-fermion (SF), modified-spin-wave (MSW), quantum Monte Carlo (QMC), and numerical-diagonalization (Exact) calculations of the ground-state energy (the left) and the excitation gap immediately above the ground state (the right) for the bond-alternating dimerized chain, where $L = 2N$ is the number of spins.

Fig. 2. The spinless-fermion (SF), modified-spin-wave (MSW), and quantum Monte Carlo (QMC) calculations of the specific heat (the upper three) and the magnetic susceptibility (the lower three) as functions of temperature for the bond-alternating dimerized chain, where $L = 2N$ is the number of spins.

The power-law prefactor to the activation-type temperature dependence, which can hardly be extracted from numerical findings, is essential in estimating the spin gap experimentally.

Next we consider the total magnetization as a function of an applied field and temperature. We compare the fermionic description of magnetization curves with numerical findings in Fig. 3. The spinless fermions again work very well. Quantum Monte Carlo sampling becomes less and less feasible with decreasing temperature, while we have no difficulty in calculating Eq. (11) even at zero temperature. The ground-state magnetization turns out to behave as $M \propto (H - H_c)^{1/2}$ near the critical field $g\mu_B H_c = E_{\text{gap}}$ [97]. Magnetization plateaux of multi-leg spin ladders [49] and mixed spin chains [98] are also well interpreted in terms of the spinless fermions. On the contrary, in the modified spin-wave theory, the number of sublattice bosons

are kept constant and therefore we have no quantitative information on the uniform magnetization as well as the staggered one. Though the Schwinger-boson mean-field theory [51,54,99,100], which consists of a rotationally invariant bosonic representation, still works with an applied field and/or existent anisotropy [52,101,102] to a certain extent but rapidly loses its validity with increasing temperature [54].

Thus and thus, we are fully convinced that the spinless fermions are superior to the modified spin waves in investigating quantum and thermal properties of spin-gapped antiferromagnets. Lastly in this section, we calculate the nuclear spin-lattice relaxation rate $1/T_1$ in terms of the spinless fermions in an attempt to stimulate further experimental interest in this system. If we again employ the approximate dispersion (25) at moderate fields and temperatures,
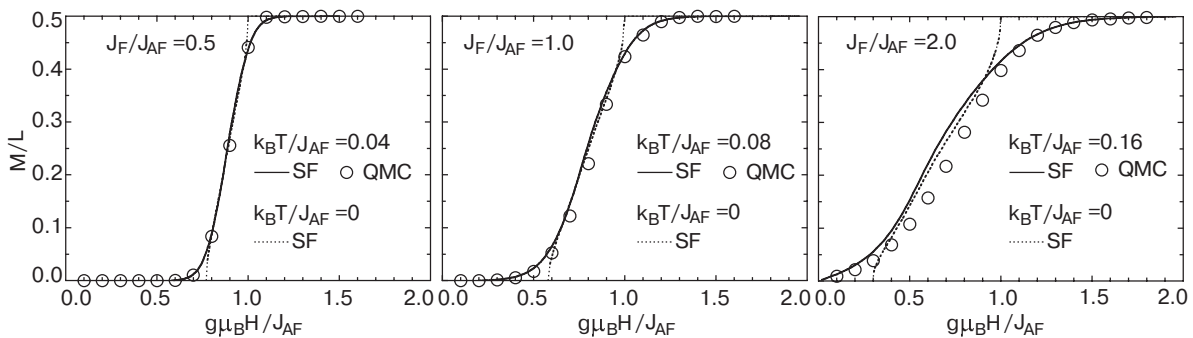

Fig. 3. The spinless-fermion and quantum Monte Carlo calculations of magnetization curves for the bond-alternating dimerized chain, where L = 2N is the number of spins.

$k_B T \ll E_{\text{gap}} - g\mu_B H$, Eq. (14) can be further calculated analytically as

$$\frac{1}{T_1} \simeq \frac{(g\mu_B \hbar \gamma_N)^2}{2\pi \hbar v J_{AF}} (A + B)^2 e^{-E_{\text{gap}}/k_B T} \times$$

$$\times \cosh \frac{g\mu_B H}{k_B T} K_0\left(\frac{\hbar\omega_N}{2k_B T}\right), \qquad (28)$$

where K_0 is the modified Bessel function of the second kind and behaves as $K_0(x) \simeq \ln 2 - \gamma - \ln x$ for $0 < x \ll 1$ with γ being Euler's constant. Considering the significant difference between the electronic and nuclear energy scales ($\hbar\omega_N \lesssim 10^{-5} J$), there usually holds the condition $\hbar\omega_N \ll k_B T$. At low temperatures, $1/T_1$ also exhibits an increase of the activation type but with logarithmic correction, which is much weaker than the power correction in the case of the susceptibility. Such a pure spin-gap-activated temperature dependence of $1/T_1$, which is shown in Fig. 4, should indeed be observed experimentally, unless magnetic impurities mask the intrinsic properties. Equation (28) further reveals a unique field dependence of $1/T_1$: *With increasing field, $1/T_1$ first decreases logarithmically and then increases exponentially*, which is visualized in Fig. 4. The initial logarithmic behavior comes from the Van Hove singularity peculiar to one-dimensional energy spectra and may arise from a nonlinear dispersion relation at the band bottom in more general. Therefore, besides spin-gapped antiferromagnets, one-dimensional ferromagnets and ferrimagnets may exhibit a similar field dependence [69,72,103,104]. Relaxation-time measurements on spin-gapped chain antiferromagnets such as IPACuCl3 and (4-BzpipdH)CuCl$_3$ are strongly encouraged.

4. Bond-alternating ferrimagnetic chain

Before closing our comparative study, we briefly mention a bond-alternating but ferrimagnetic chain calculated within the same schemes. We take another interest in the ferromagnetic−ferromagnetic−antiferromagnetic−antiferromagnetic bond-tetrameric spin-1/2 Heisenberg chain, whose Hamiltonian is given by

$$\mathcal{H} = \sum_{n=1}^{N} [J_{AF}(\mathbf{S}_{4n-3}\cdot\mathbf{S}_{4n-2} + \mathbf{S}_{4n-2}\cdot\mathbf{S}_{4n-1}) - \\ -J_F(\mathbf{S}_{4n-1}\cdot\mathbf{S}_{4n} + \mathbf{S}_{4n}\cdot\mathbf{S}_{4n+1})]. \qquad (29)$$

Cu(3-Clpy)$_2$(N$_3$)$_2$ (3-Clpy = 3-chloropyridine = C$_5$ClH$_4$N) [105] is well described by this Hamiltonian [106] and behaves as if it is a ferrimagnet of alternating spins 3/2 and 1/2 [68]. In the conventional spin-wave scheme, the spin deviations in each sublattice, $\langle a_{\tau:n}^\dagger a_{\tau:n}\rangle$ and $\langle b_{\tau:n}^\dagger b_{\tau:n}\rangle$, diverge in the antiferromagnetic ground state but stay finite in the ferrimagnetic one. Without quantum divergence of the sublattice magnetization, it is not necessary to diagonalize the effective Hamiltonian (29). In an attempt to keep the dispersion relations free from temperature, we may simply diagonalise the original Hamiltonian (18) and then introduce a Lagrange multiplier so as to minimize the free energy [54]. For ferrimagnets such an idea is much superior to the original antiferromagnetic modified spin-wave scheme [56–58].

Figure 5 shows the thus-modified spin-wave calculations as well as the Hartree−Fock calculations in terms of the spinless fermions in comparison with numerical findings. The ferrimagnetic modified spin waves work very well, contrasting with the antiferromagnetic ones. They reproduce the Schottky-type
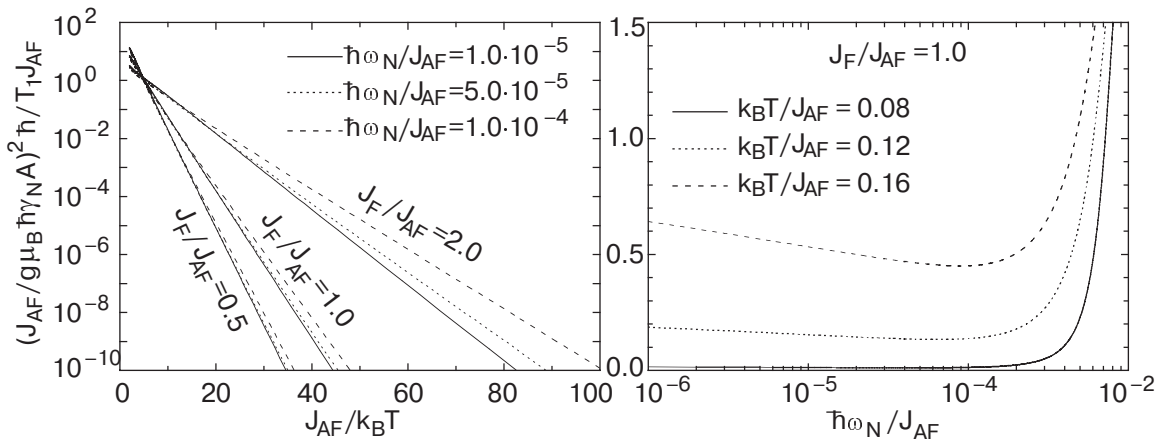

Fig. 4. The spinless-fermion alculations of the nuclear spin-lattice relaxation rate as a function of temperature (the left) and an applied magnetic field (the right) for the bond-alternating dimerized chain.
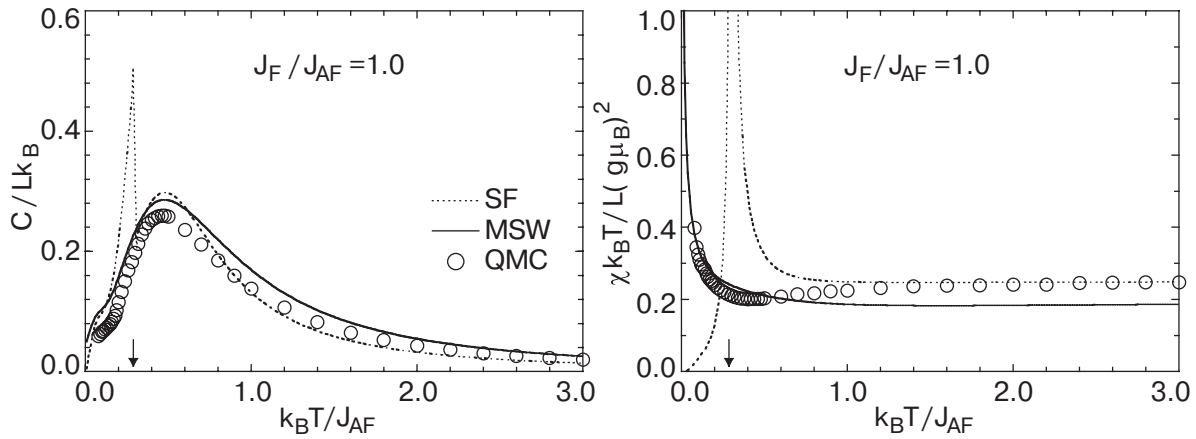
Fig. 5. The spinless-fermion, modified-spin-wave, and quantum Monte Carlo calculations of the specific heat (the left) and the magnetic susceptibility (the right) as functions of temperature for the bond-alternating tetramerized chain, where $L \equiv 4N$ is the number of spins. The Hartree−Fock fermions encounter a paramagnetic-to-Néel-ordered phase transition with decreasing temperature and the transition temperature is indicated by arrows.

peak of the specific heat and the ferrimagnetic minimum of the susceptibility-temperature product at intermediate temperatures. Although the modified spin-wave description of the antiferromagnetic increase of χT is somewhat moderate, it converges into the paramagnetic value $S(S+1)/3$ at high temperatures. The description is more and more refined with decreasing temperature and is expected to be accurate at sufficiently low temperatures [67]. The $T^{1/2}$-initial behavior of C and the T^{-2}-diverging behavior of χ are both correctly reproduced [66]. Besides static properties, T_1 measurements [107] on a ferrimagnetic chain compound NiCu($C_7H_6N_2O_6$)(H_2O_3)·$2H_2O$ was elaborately interpreted in terms of the modified spin waves [108].

On the other hand, the spinless fermions misread the low-temperature properties of ferrimagnetic chains. A fatally weak point of their description is the onset of a Néel-ordered state. With increasing J_F, the transition temperature T_c goes up and the applicability of the Hartree−Fock fermions is reduced. Indeed the fermionic description is not so bad away upward from T_c, but it is much less complementary to numerical tools in ferrimagnetic systems.

5. Summary

We have comparatively discussed fermionic and bosonic descriptions of the bond-dimeric Heisenberg chain as an example of spin-gapped antiferromagnets. The fermionic language is based on the Jordan−Wigner spinless fermions within the Hartree−Fock approximation, while the bosonic formulation consists of constraining the Holstein−Primakoff bosons to restore the sublattice symmetry. The spinless fermions well describe both ground-state and finite-temperature pro-

perties. The zero-field specific heat and magnetic susceptibility behave as

$$C \propto (k_B T)^{-3/2} e^{-E_{\mathrm{gap}}/k_B T}$$

and

$$\chi \propto (k_B T)^{-1/2} e^{-E_{\mathrm{gap}}/k_B T},$$

respectively, at sufficiently low temperatures, while the relaxation rate as

$$1/T_1 \propto e^{-E_{\mathrm{gap}}/k_B T} \cosh(g\mu_B H/k_B T)[0.80908 - \\ - \ln(\hbar\omega_N/k_B T)]$$

at moderately low temperatures and fields. On the other hand, the modified spin waves give much poorer findings. In particular, they significantly underestimate the spin gap and fail to reproduce the Schottky-type peak of the specific heat. The same schemes have further been applied to the bond-tetrameric ferrimagnetic chain, where the modified spin waves work very well and are superior to the spinless fermions both qualitatively and quantitatively. The fermionic language is useful in describing disordered ground states and their excitations, whereas the bosonic one in depicting ordered ground states and their fluctuations.

The modified spin-wave theory is fully applicable to higher-spin systems. The Jordan−Wigner transformation can also be generalized to higher-spin systems [109], where spin-1 chains, for instance, are mapped onto an extended $t-J$ model of strongly correlated electrons. However, the *double-graded* Hubbard operators such as $\tilde{c}_{n,\uparrow} \equiv (1 - c_{n,\downarrow}^\dagger c_{n,\downarrow})c_{n,\uparrow}$ demand that we should treat the fermion and boson degrees of freedom in the same footing [99,110−112]. The present

naive fermionic representation is highly successful for spin-1/2 gapped antiferromagnets, including various bond-alternating and/or coupled chains. It is complementary to numerical tools especially at low temperatures and allows us to readily infer both static and dynamic properties of spin-gapped antiferromagnets

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