Bosonic representation of one-dimensional Heisenberg ferrimagnets

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We present a comparative study of bosonic languages to describe one-dimensional Heisenberg ferrimagnets. The ferrimagnetic Schwinger-boson mean-field theory demonstrated by Wu *et al.*, the antiferromagnetic modified spin-wave theory designed by Takahashi, and its ferrimagnetic variant proposed by Yamamoto *et al.* are employed to calculate the energy structure and the thermodynamics of various ferrimagnets. A modified spin-wave scheme, which introduces a Lagrange multiplier keeping the native energy structure free from temperature and thus differs from the original Takahashi scheme, is particularly stressed as a useful tool to investigate one-dimensional quantum ferrimagnetism. The antiferromagnetic limit of these descriptions is also considered.

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I. INTRODUCTION

Significant efforts have been devoted to synthesizing low-dimensional ferrimagnets and understanding their quantum behavior in recent years. The first example of one-dimensional ferrimagnets, $MnCu(S_2C_2O_2)_2(H_2O)_3 \cdot 4.5H_2O$, was synthesized by Gleizes and Verdaguer[1] and followed by a series of ordered bimetallic chain compounds[2] in an attempt to design molecule-based ferromagnets.[3] Caneschi *et al.*[4] demonstrated another approach to alternating-spin chains hybridizing manganese complexes and nitronyl nitroxide radicals. The inorganic-organic hybrid strategy realized more complicated alignments of mixed spins.[5] There also exists an attempt at stacking novel triradicals into a purely organic ferrimagnet.[6] Monospin chains can be ferrimagnetic with polymerized exchange interactions. An example of such ferrimagnets is the ferromagnetic-antiferromagnetic bond-alternating copper tetramer chain compound $Cu(C_5H_4NCl)_2(N_3)_2$.[7] The trimeric intertwining double-chain compound $Ca_3Cu_3(PO_4)_4$ (Ref. 8) is another solution to homometallic one-dimensional ferrimagnets, where the noncompensation of sublattice magnetizations is of topological origin. Besides one-dimensional ferrimagnets, metal-ion magnetic clusters such as $[Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4]$ (Ref. 9) and $[Fe_8(N_3C_6H_{15})_6O_2(OH)_{12}]^{8+}$,[10] for which resonant magnetization tunneling[11–14] was observed, are also worth mentioning as zero-dimensional ferrimagnets.

The discovery of ordered bimetallic chain compounds stimulated extensive theoretical interest in (quasi-)one-dimensional quantum ferrimagnets. Early efforts[15] were devoted to numerically diagonalizing alternating-spin Heisenberg chains. Numerical diagonalization, combined with the Lanczos algorithm[16,17] and a scaling technique,[18] further contributed to studying modern topics such as phase transitions of the Kosterlitz-Thouless type[17,19] and quantized magnetization plateaux.[20,21] Alternating-spin chains were further investigated by density-matrix renormalization-group[22,23] and quantum Monte Carlo[24,25] methods in an attempt to illuminate dual features of ferrimagnetic excitations. More general mixed-spin chains were analyzed via the nonlinear σ model[26] with particular emphasis on the competition between massive and massless phases. Quasi-one-dimensional mixed-spin systems[27,28] were also investigated in order to explain the inelastic-neutron-scattering findings[29,30] for the rare-earth nickelates R_2BaNiO_5.

In order to complement numerical tools and to achieve further understanding of the magnetic double structure of ferrimagnetism, several authors have recently begun to construct bosonic theories of low-dimensional quantum ferrimagnets. The conventional spin-wave description of the ground-state properties,[22,31–33] a modified spin-wave scheme for the low-temperature properties,[34] and the Schwinger-boson representation of the low-energy structure[35] and the thermodynamics,[36] they all reveal the potential of bosonic languages for various ferrimagnetic systems. However, considering the global argument and total understanding over the bosonic theory of ferromagnets and antiferromagnets,[37–47] ferrimagnets are still undeveloped in this context especially in one dimension. In such circumstances, we represent one-dimensional Heisenberg ferrimagnets in terms of the Schwinger bosons and the Holstein-Primakoff spin waves. Based on a mean-field ansatz, the local constraints on the Schwinger bosons are relaxed and imposed only on the average. The conventional antiferromagnetic spin-wave formalism[48,49] is modified, on the one hand following the Takahashi scheme[37,38] which was originally proposed for ferromagnets, while on the other hand introducing a slightly different strategy.[50] The Schwinger bosons and the modified spin waves both interpret the low-energy properties fairly well identifying the ferrimagnetic long-range order with a Bose condensation, while the two languages are qualitatively distinguished in describing the thermodynamics. We demonstrate that the modified spin-wave scheme is much better than the others at describing one-dimensional ferrimagnets.

II. FORMALISM

A practical model for one-dimensional ferrimagnets is two kinds of spins, S and s $(S>s)$, alternating on a ring with antiferromagnetic exchange coupling between nearest neighbors, as described by the Hamiltonian,

$$\mathcal{H}=J\sum_{n=1}^{N}(\boldsymbol{S}_n\cdot\boldsymbol{s}_{n-1}+\boldsymbol{s}_n\cdot\boldsymbol{S}_n). \tag{2.1}$$

Hereafter, the distance between neighboring spins is represented by a. The simplest case, $(S,s)=(1,\frac{1}{2})$, has so far been discussed fairly well using the matrix-product formalism,[51] a modified spin-wave scheme,[50] the Schwinger-boson representation,[36] and modern numerical techniques.[22–25] We make further explorations into higher-spin systems and develop the analytic argument in more detail.

A. Schwinger-boson mean-field theory

Let us describe each spin variable in terms of two kinds of bosons as

$$S_n^+=a_n^\dagger a_n\,,\quad S_n^z=\tfrac{1}{2}(a_{n+}^\dagger a_n-a_{n-}^\dagger a_{n-}),$$

$$s_n^+=b_{n+}^\dagger b_{n-}\,,\quad s_n^z=\tfrac{1}{2}(b_{n+}^\dagger b_{n+}-b_{n-}^\dagger b_{n-}), \tag{2.2}$$

where the constraints

$$\sum_{\sigma=\pm}a_{n\sigma}^\dagger a_{n\sigma}=2S,\quad \sum_{\sigma=\pm}b_{n\sigma}^\dagger b_{n\sigma}=2s, \tag{2.3}$$

are imposed on the bosons. Relaxing the constraints (2.3) as

$$\sum_{n=1}^{N}\sum_{\sigma=\pm}a_{n\sigma}^\dagger a_{n\sigma}=2NS,\quad \sum_{n=1}^{N}\sum_{\sigma=\pm}b_{n\sigma}^\dagger b_{n\sigma}=2Ns, \tag{2.4}$$

and assuming the thermal average of the short-range antiferromagnetic order to be uniform and static as

$$\langle a_{n+}b_{\tilde{n}-}-a_{n-}b_{\tilde{n}+}\rangle_T=2\Omega, \tag{2.5}$$

with $\tilde{n}=n,n-1$, we obtain the mean-field Hamiltonian in the momentum space as

$$\begin{aligned}\mathcal{H}_{\rm MF}=&2NJSs+4NJ\Omega^2-4NJ(\lambda S+\mu s)\\&-2J\Omega\sum_k\cos ak(a_{k+}b_{k-}-a_{k-}b_{k+}+\text{H.c.})\\&+2J\sum_k\sum_{\sigma=\pm}(\lambda a_{k\sigma}^\dagger a_{k\sigma}+\mu b_{k\sigma}^\dagger b_{k\sigma}),\end{aligned} \tag{2.6}$$

where λ and μ are the Lagrange multipliers due to the constraints (2.4). Via the Bogoliubov transformation

$$a_{k\sigma}=\alpha_{k\sigma}\cosh\theta_k+\sigma\beta_{k-\sigma}^\dagger\sinh\theta_k\,,$$

$$b_{k\sigma}=\beta_{k\sigma}\cosh\theta_k-\sigma\alpha_{k-\sigma}^\dagger\sinh\theta_k\,, \tag{2.7}$$

with

$$\tanh 2\theta_k=\frac{2\Omega\cos ak}{\lambda+\mu}, \tag{2.8}$$

the Hamiltonian (2.6) is diagonalized as

$$\begin{aligned}\mathcal{H}_{\rm MF}=&2NJSs+4NJ\Omega^2-2NJ\lambda(2S+1)-2NJ\mu(2s+1)\\&+2J\sum_k\omega_k+J\sum_k\sum_{\sigma=\pm}(\omega_{k\sigma}^-\alpha_{k\sigma}^\dagger\alpha_{k\sigma}+\omega_{k\sigma}^+\beta_{k\sigma}^\dagger\beta_{k\sigma}),\end{aligned} \tag{2.9}$$

where

$$\omega_{k\sigma}^\pm\equiv\omega_k^\pm=\omega_k\pm(\mu-\lambda),$$

$$\omega_k=\sqrt{(\lambda+\mu)^2-4\Omega^2\cos^2 ak}. \tag{2.10}$$

λ, μ, and Ω are determined through a set of equations

$$\sum_k(\overline{n_{k\sigma}^-}\cosh^2\theta_k+\overline{n_{k\sigma}^+}\sinh^2\theta_k+\sinh^2\theta_k)=NS, \tag{2.11}$$

$$\sum_k(\overline{n_{k\sigma}^-}\sinh^2\theta_k+\overline{n_{k\sigma}^+}\cosh^2\theta_k+\sinh^2\theta_k)=Ns, \tag{2.12}$$

$$\sum_k(\overline{n_{k\sigma}^-}+\overline{n_{k\sigma}^+}+1)\cosh\theta_k\sinh\theta_k=N\Omega, \tag{2.13}$$

where the thermal distribution functions $\overline{n_{k\sigma}^-}\equiv\langle\alpha_{k\sigma}^\dagger\alpha_{k\sigma}\rangle_T$ and $\overline{n_{k\sigma}^+}\equiv\langle\beta_{k\sigma}^\dagger\beta_{k\sigma}\rangle_T$ are required to minimize the free energy and given by

$$\overline{n_{k\sigma}^\pm}=\frac{1}{e^{\omega_{k\sigma}^\pm/k_{\rm B}T}-1}. \tag{2.14}$$

The magnetic susceptibility is expressed as

$$\chi=\frac{(g\mu_{\rm B})^2}{4k_{\rm B}T}\sum_k\sum_{\tau=\pm}\sum_{\sigma=\uparrow,\downarrow}\overline{n_{k\sigma}^\tau}(\overline{n_{k\sigma}^\tau}+1), \tag{2.15}$$

where we have set the g factors of spins $\boldsymbol{S}$ and $\boldsymbol{s}$ both equal to g. The internal energy should be given by

$$E=\tfrac{1}{2}(E_{\rm MF}+2NJSs)-2NJSs, \tag{2.16}$$

where

$$\begin{aligned}E_{\rm MF}=&2NJSs+4NJ\Omega^2-2NJ(2\lambda S+2\mu s+\lambda+\mu)\\&+2J\sum_k\omega_k+J\sum_k\sum_{\tau=\pm}\sum_{\sigma=\pm}\overline{n_{k\sigma}^\tau}\omega_{k\sigma}^\tau.\end{aligned} \tag{2.17}$$

Arovas and Auerbach[44] pointed out that relaxing the original constraints (2.3) into Eq. (2.4) leads to double counting the number of independent boson degrees of freedom. Therefore, in Eq. (2.16), we have corrected the mean-field artifact reducing the overestimated quantum fluctuation.

B. Modified spin-wave theory: Takahashi scheme

Next we consider a single-component bosonic representation of each spin variable at the cost of the rotational symmetry. We start from the Holstein-Primakoff transformation

$$S_n^+ = \sqrt{2S - a_n^\dagger a_n}\, a_n, \quad S_n^z = S - a_n^\dagger a_n,$$

$$s_n^+ = b_n^\dagger \sqrt{2s - b_n^\dagger b_n}, \quad s_n^z = -s + b_n^\dagger b_n. \tag{2.18}$$

Treating S and s as $O(S) = O(s)$, we can expand the Hamiltonian with respect to $1/S$ as

$$\mathcal{H} = -2NJSs + E_1 + E_0 + \mathcal{H}_1 + \mathcal{H}_0 + O(S^{-1}), \tag{2.19}$$

where E_i and $\mathcal{H}_i$ give the $O(S^i)$ quantum corrections to the ground-state energy and the dispersion relations, respectively. Via the Bogoliubov transformation

$$a_k = \alpha_k \cosh\theta_k - \beta_k^\dagger \sinh\theta_k,$$

$$b_k = \beta_k \cosh\theta_k - \alpha_k^\dagger \sinh\theta_k, \tag{2.20}$$

they are written as

$$E_1 = -2NJ[2\sqrt{Ss}\,\Gamma - (S+s)\Lambda], \tag{2.21a}$$

$$E_0 = -2NJ\left[\Gamma^2 + \Lambda^2 - \left(\sqrt{\frac{S}{s}} + \sqrt{\frac{s}{S}}\right)\Gamma\Lambda\right], \tag{2.21b}$$

$$\mathcal{H}_i = J\sum_k [\omega_i^-(k)\alpha_k^\dagger\alpha_k + \omega_i^+(k)\beta_k^\dagger\beta_k + \gamma_i(k)(\alpha_k\beta_k + \alpha_k^\dagger\beta_k^\dagger)], \tag{2.22}$$

where

$$\Gamma = \frac{1}{2N}\sum_k \cos ak \sinh 2\theta_k, \tag{2.23}$$

$$\Lambda = \frac{1}{2N}\sum_k (\cosh 2\theta_k - 1), \tag{2.24}$$

$$\omega_1^\pm(k) = (S+s)\cosh 2\theta_k - 2\sqrt{Ss}\cos ak \sinh 2\theta_k \pm (S-s) \equiv \omega_k \pm (S-s), \tag{2.25a}$$

$$\omega_0^\pm(k) = \left[\left(\sqrt{\frac{S}{s}} + \sqrt{\frac{s}{S}}\right)\Gamma - 2\Lambda\right]\cosh 2\theta_k - \left[2\Gamma - \left(\sqrt{\frac{S}{s}} + \sqrt{\frac{s}{S}}\right)\Lambda\right]\cos ak \sinh 2\theta_k \pm \left(\sqrt{\frac{S}{s}} - \sqrt{\frac{s}{S}}\right), \tag{2.25b}$$

$$\gamma_1(k) = 2\sqrt{Ss}\cos ak \cosh 2\theta_k - (S+s)\sinh 2\theta_k, \tag{2.26a}$$

$$\gamma_0(k) = \left[2\Gamma - \left(\sqrt{\frac{S}{s}} + \sqrt{\frac{s}{S}}\right)\Lambda\right]\cos ak \cosh 2\theta_k - \left[\left(\sqrt{\frac{S}{s}} + \sqrt{\frac{s}{S}}\right)\Gamma - 2\Lambda\right]\sinh 2\theta_k. \tag{2.26b}$$

$\mathcal{H}_0$ originally consists of quartic bosonic terms and has been decomposed by means of the Wick theorem. The residual two-body interactions have been neglected so as to keep the ferromagnetic excitation branch gapless.

The conventional spin-wave scheme naively diagonalize the Hamiltonian (2.19) and ends up with the number of bosons diverging with increasing temperature. In order to suppress this thermal divergence, Takahashi[38] considered optimizing the bosonic distribution functions under zero magnetization and obtained an excellent description of the low-temperature thermodynamics for low-dimensional Heisenberg ferromagnets. For ferrimagnets, this idea is still useful[34,50] but never applies away from the low-temperature region as it is. The zero-magnetization constraint plays a role of keeping the number of bosons finite under ferromagnetic interactions but does not work so under antiferromagnetic interactions. Takahashi[38] and Hirsch *et al.*[39] proposed constraining the staggered magnetization, instead of the uniform magnetization, to be zero as the antiferromagnetic version of the modified spin-wave theory. Their scheme was applied to extensive antiferromagnets in both two[38,39,52–55] and one[40,41] dimensions. The conventional spin-wave procedure assumes that spins on one sublattice point predominantly up, while those on the other predominantly down. The modified spin-wave treatment restores the sublattice symmetry. We consider the naivest generalization of the antiferromagnetic modified spin-wave scheme to ferrimagnets.

The constraint of zero staggered magnetization reads

$$\sum_n (a_n^\dagger a_n + b_n^\dagger b_n) = N(S+s). \tag{2.27}$$

In order to enforce this condition, we first introduce a Lagrange multiplier and diagonalize the effective Hamiltonian

$$\widetilde{\mathcal{H}} = \mathcal{H} + 2J\nu\sum_n (a_n^\dagger a_n + b_n^\dagger b_n). \tag{2.28}$$

Then the ground-state energy and the dispersion relations are obtained as

$$E_{\mathrm{g}} = -2NJSs + \widetilde{E}_1, \quad \widetilde{E}_1 = E_1 + 4NJ\Lambda\nu, \tag{2.29}$$

$$\omega_k^\pm = \widetilde{\omega}_1^\pm(k), \quad \widetilde{\omega}_1^\pm(k) = \omega_1^\pm(k) + 2\nu\cosh 2\theta_k, \tag{2.30}$$

keeping only the bilinear terms and as

$$E_{\mathrm{g}} = -2NJSs + \widetilde{E}_1 + E_0, \tag{2.31}$$

$$\omega_k^\pm = \widetilde{\omega}_1^\pm(k) + \omega_0^\pm(k), \tag{2.32}$$

considering the $O(S^0)$ interactions as well. In terms of the spin-wave distribution functions

$$\bar{n}_k^\pm = \frac{1}{e^{\omega_k^\pm/k_{\mathrm{B}}T} - 1}, \tag{2.33}$$

the internal energy and the magnetic susceptibility are expressed as[56]

$$E=E_{\mathrm{g}}+\sum_k \sum_{\tau=\pm} \overline{n_k^\tau}\omega_k^\tau, \qquad (2.34)$$

$$\chi=\frac{(g\mu_{\mathrm{B}})^2}{3k_{\mathrm{B}}T}\sum_k \sum_{\tau=\pm} \overline{n_k^\tau}(\overline{n_k^\tau}+1). \qquad (2.35)$$

θ_k, defining the Bogoliubov transformation (2.20), is determined through

$$\gamma_1(k)-2\nu\sinh 2\theta_k \equiv \tilde{\gamma}_1(k)=0, \qquad (2.36)$$

provided we treat $\mathcal{H}_0$ as a perturbation to $\mathcal{H}_1$.

C. Modified spin-wave theory: A different scheme

Although the Takahashi scheme overcomes the difficulty of sublattice magnetizations diverging thermally, the obtained thermodynamics is still far from satisfactory (see Fig. 2 later on). Within the conventional spin-wave theory, the quantum spin reduction, that is, the quantum fluctuation of the ground-state sublattice magnetization per unit cell, reads

$$\langle a_n^\dagger a_n\rangle_{T=0}=\langle b_n^\dagger b_n\rangle_{T=0}\equiv\delta$$
$$=\int_0^\pi \frac{S+s}{\sqrt{(S-s)^2+4Ss\sin^2(k/2)}}\frac{\mathrm{d}k}{2\pi}-\frac{1}{2}, \qquad (2.37)$$

and diverges at $S=s$. The Takahashi scheme settles this *quantum divergence* as well as the *thermal divergence*. However, the number of bosons does not diverge in the ferrimagnetic ground state. Without quantum divergence, it is not necessary to modify the dispersion relations (2.25a) into the temperature-dependent form (2.30). While the thermodynamics should be modified, the quantum mechanics may be left as it is.

Such an idea leads to the Bogoliubov transformation free from temperature replacing Eq. (2.36) by $\gamma_1(k)=0$, that is,

$$\tanh 2\theta_k=\frac{2\sqrt{Ss}\cos ak}{S+s}. \qquad (2.38)$$

The ground-state energy and the dispersion relations are simply given by

$$E_{\mathrm{g}}=-2NJSs+E_1, \quad \omega_k^\pm=\omega_1^\pm(k), \qquad (2.39)$$

within the up-to-$O(S^1)$ treatment and by

$$E_{\mathrm{g}}=-2NJSs+E_1+E_0, \quad \omega_k^\pm=\omega_1^\pm(k)+\omega_0^\pm(k), \qquad (2.40)$$

in the up-to-$O(S^0)$ treatment. They are nothing but the $T=0$ findings in the Takahashi scheme.

At finite temperatures we replace $\alpha_k^\dagger\alpha_k$ and $\beta_k^\dagger\beta_k$ in the spin-wave Hamiltonian (2.22) by

$$\overline{n_k^\mp}\equiv\sum_{n^-,n^+=0}^\infty n^\mp P_k(n^-,n^+), \qquad (2.41)$$

where $P_k(n^-,n^+)$ is the probability of n^- ferromagnetic and n^+ antiferromagnetic spin waves appearing in the k-momentum state and satisfies

$$\sum_{n^-,n^+} P_k(n^-,n^+)=1 \qquad (2.42)$$

for all k's. Then the free energy is written as

$$F=E_{\mathrm{g}}+J\sum_k \sum_{n^-,n^+} P_k(n^-,n^+)\sum_{\tau=\pm} n^\tau\omega_k^\tau$$
$$+k_{\mathrm{B}}T\sum_k \sum_{n^-,n^+} P_k(n^-,n^+)\ln P_k(n^-,n^+). \qquad (2.43)$$

We minimize the free energy with respect to $P_k(n^-,n^+)$ enforcing a condition

$$\langle S_n^z-s_n^z\rangle_T+2\delta\equiv\langle :S_n^z-s_n^z:\rangle_T$$
$$=S+s-\frac{S+s}{N}\sum_k \sum_{\tau=\pm}\frac{\overline{n_k^\tau}}{\omega_k}=0, \qquad (2.44)$$

as well as the trivial constraints (2.42). In the second-side compact expression, the normal ordering is taken with respect to both operators α and β. Equation (2.44) claims that the thermal fluctuation $(S+s)\Sigma_k(n_k^-+n_k^+)/\omega_k$ should cancel the *full*, or *classical*, Néel order $(S+s)N$ rather than the *quantum-mechanically reduced* one $(S+s-2\delta)N$. Without consideration of the quantum fluctuation 2δ, which is absent from ferromagnets but peculiar to ferrimagnets, the present

TABLE I. The Schwinger-boson (SB), linear-modified-spin-wave (LMSW), perturbational interacting-modified-spin-wave (PIMSW), and numerical diagonalization (exact) calculations of the ground-state energy E_{g} and the zero-temperature antiferromagnetic excitation gap Δ_0 for the spin-(S,s) ferrimagnetic Heisenberg chains.

	$(S,s)=(1,\frac{1}{2})$		$(S,s)=(\frac{3}{2},\frac{1}{2})$		$(S,s)=(\frac{3}{2},1)$	
Approach	E_{g}/NJ	Δ_0/J	E_{g}/NJ	Δ_0/J	E_{g}/NJ	Δ_0/J
SB	−1.45525	1.77804	−1.96755	2.84973	−3.86270	1.62152
LMSW	−1.43646	1	−1.95804	2	−3.82807	1
PIMSW	−1.46084	1.67556	−1.96983	2.80253	−3.86758	1.52139
Exact	−1.4541(1)	1.759(1)	−1.9672(1)	2.842(1)	−3.861(1)	1.615(5)

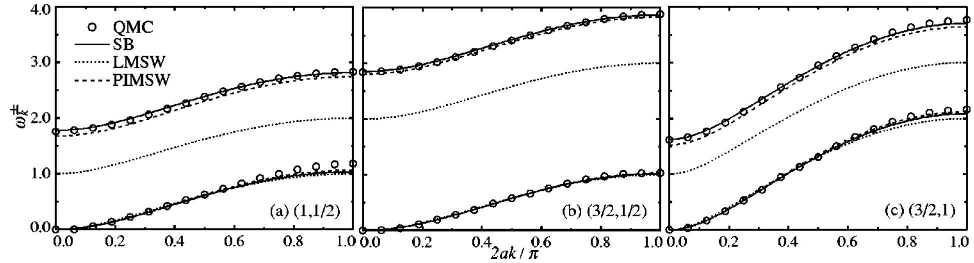

FIG. 1. The Schwinger-boson (SB), linear-modified-spin-wave (LMSW), perturbational interacting-modified-spin-wave (PIMSW), and quantum Monte Carlo (QMC) calculations of the dispersion relations of the ferromagnetic (ω_k^-) and antiferromagnetic (ω_k^+) elementary excitations for the spin-(S,s) ferrimagnetic Heisenberg chains at zero temperature.

scheme breaks even the conventional spin-wave achievement at low temperatures. Numerically solving the thermodynamic Bethe-Ansatz equations, Takahashi and Yamada[57] suggested that the conventional spin-wave theory correctly gives the low-temperature leading term of the specific heat. Both the Takahashi scheme with Eq. (2.27) and the different scheme with Eq. (2.44) indeed keep unchanged the conventional spin-wave findings

$$\frac{C}{Nk_{\mathrm{B}}} \sim \frac{3}{4}\sqrt{\frac{S-s}{Ss}}\frac{\zeta(\frac{3}{2})}{\sqrt{2\pi}}t^{1/2}\,(T\to 0), \tag{2.45}$$

where $t=k_{\mathrm{B}}T/J$ within the up-to-$O(S^1)$ treatment, while $t=k_{\mathrm{B}}T/\gamma J$ with $\gamma=1+\Gamma/\sqrt{Ss}-(S+s)\Lambda/Ss$ in the up-to-$O(S^0)$ treatment. The conventional spin-wave approach gives no quantitative information on the magnetic susceptibility, whereas the modified theory reveals

$$\frac{\chi J}{N(g\mu_{\mathrm{B}})^2} \sim \frac{Ss(S-s)^2}{3}t^{-2}\,(T\to 0). \tag{2.46}$$

In terms of the optimum distribution functions

$$\overline{n_k^{\pm}} = \frac{1}{\mathrm{e}^{[J\omega_k^{\pm}-\nu(S+s)/\omega_k]/k_{\mathrm{B}}T}-1}, \tag{2.47}$$

the free energy at the thermal equilibrium is written as

$$F=E_{\mathrm{g}}+\nu(S+s)N-k_{\mathrm{B}}T\sum_k\sum_{\tau=\pm}\ln(1+\overline{n_k^{\tau}}), \tag{2.48}$$

where ν is the Lagrange multiplier due to the constraint (2.44).

III. RESULTS

First we calculate the ground-state energy E_g and the antiferromagnetic excitation gap $\omega_{k=0}^{+}$ and compare them with numerical findings in Table I. At $T=0$, the Takahashi scheme and the different scheme both give $\nu=0$ and lead to the conventional spin-wave findings. Higher-order spin-wave calculation[32] of the ground-state properties is feasible, while more sophisticated series-expansion technique[58] has recently been proposed. We are fully convinced that the spin-wave treatment better works for larger spins. Table I further shows that the spin-wave approach is better justified with increasing S/s as well as Ss, which is because the quantity $S-s$ fills the role of suppressing the divergence in Eq. (2.37). On the other hand, the Schwinger-boson approach constantly gives highly precise estimates of the low-energy properties. Figure 1 further demonstrates that the Schwinger-boson mean-field theory is highly successful in describing the low-lying excitations. Both the bosonic languages well interpret the ferromagnetic excitations, whereas the linear spin waves considerably underestimate the antiferromagnetic excitation energies. The quantum correlation has much effect on the antiferromagnetic excitation mode and such an effect is well included into the Schwinger-boson calculation even at the mean-field level.

Next we calculate the thermodynamic properties. Figure 2 shows the temperature dependence of the specific heat. The Schwinger-boson mean-field theory is still highly successful

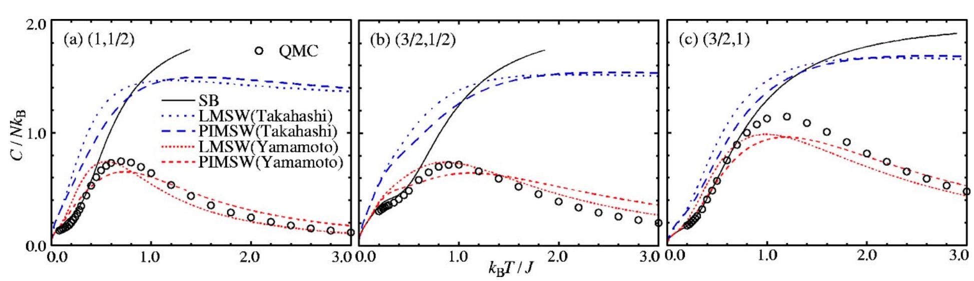

FIG. 2. (Color online) The Schwinger-boson (SB), linear-modified-spin-wave (LMSW), perturbational interacting-modified-spin-wave (PIMSW), and quantum Monte Carlo (QMC) calculations of the specific heat C as a function of temperature for the spin-(S,s) ferrimagnetic Heisenberg chains. The modified spin waves are constructed in two different ways, the Takahashi scheme (Takahashi) and the different scheme (Yamamoto).

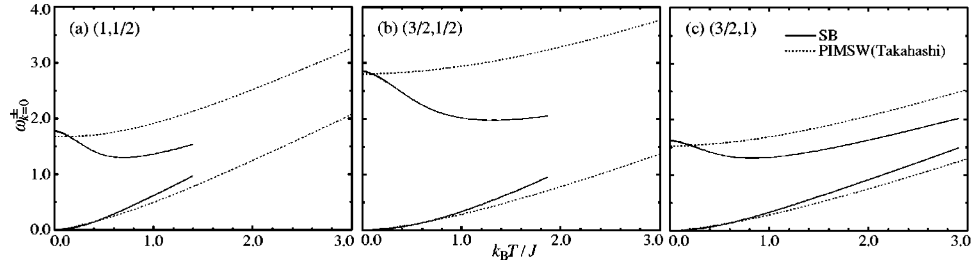

FIG. 3. The ferromagnetic ($\omega^{-}_{k=0}$) and antiferromagnetic ($\omega^{+}_{k=0}$) excitation gaps as functions of temperature for the spin-(S,s) ferrimagnetic Heisenberg chains calculated by the Schwinger bosons (SB) and the perturbationally interacting modified spin waves (PIMSW) based on the Takahashi scheme.

at low temperatures, while with increasing temperature, it rapidly breaks down failing to reproduce the Schottky-type peak. The mean-field order parameter Ω monotonically decreases with increasing temperature and reaches zero at

$$\frac{k_B T}{J} = \frac{S+s+1}{\ln(1+1/S)+\ln(1+1/s)}. \tag{3.1}$$

Above this temperature, Ω sticks at zero suggesting no antiferromagnetic correlation in the system. The onset of the paramagnetic phase at a finite temperature is a mean-field artifact and the particular temperature (3.1) is an increasing function of S and s. The modified spin-wave theory based on the Takahashi scheme also fails to describe the Schottky peak. Because of the Lagrange multiplier ν, which turns out a monotonically increasing function of temperature, the dispersion relations (2.30) lead to endlessly increasing energy and thus nonvanishing specific heat at high temperatures. Only the modified spin-wave theory based on the different scheme succeeds in interpreting the Schottky peak. Since the antiferromagnetic excitation gap is significantly improved by the inclusion of the $O(S^0)$ correlation, the interacting modified spin waves reproduce the location of the Schottky peak fairly well. Mixed-spin trimeric chain ferrimagnets have recently been synthesized[5] and their low-temperature thermal properties were well elucidated by the modified spin-wave theory.[34] However, it was unfortunate that the additional constraint was imposed on the uniform magnetization and therefore the higher-temperature properties were much less illuminated. Controlling the staggered magnetization instead based on the different scheme, we can fully investigate such polymeric chain compounds as well.

In the Schwinger representation and the modified spin-wave treatment based on the Takahashi scheme, the energy spectrum depends on temperature. Since the low-energy band structure is well reflected in the thermal behavior and can directly be observed through inelastic-neutron-scattering measurements, we investigate the ferromagnetic ($\omega^{-}_{k=0}$) and antiferromagnetic ($\omega^{+}_{k=0}$) excitation gaps as functions of temperature in Fig. 3. The Schwinger-boson mean-field theory claims that the antiferromagnetic gap should first decrease and then increase with increasing temperature, while the modified spin-wave theory predicts that the excitation energies of both modes should be monotonically increasing functions of temperature. We find a similar contrast between the two languages applied to ladder ferrimagnets.[35,59] In the case of Haldane-gap antiferromagnets, both the Schwinger-boson and modified-spin-wave[41] findings, together with the nonlinear-σ-model calculations,[60,61] commonly suggest that the Haldane gap is a simply activated function of temperature. Extensive measurements on spin-1 antiferromagnetic Heisenberg chain compounds[62–64] also report that the Haldane massive mode is shifted upward with increasing temperature. Neutron-scattering experiments on ferrimagnetic chain compounds may solve the present disagreement between the Schwinger-boson and modified-spin-wave calculations of the antiferromagnetic excitation gap as a function of temperature.

Figure 4 shows the temperature dependence of the magnetic-susceptibility-temperature product, which elucidates ferromagnetic and antiferromagnetic features coexisting in ferrimagnets.[25] χT diverges at low temperatures in a ferromagnetic fashion but approaches the high-temperature

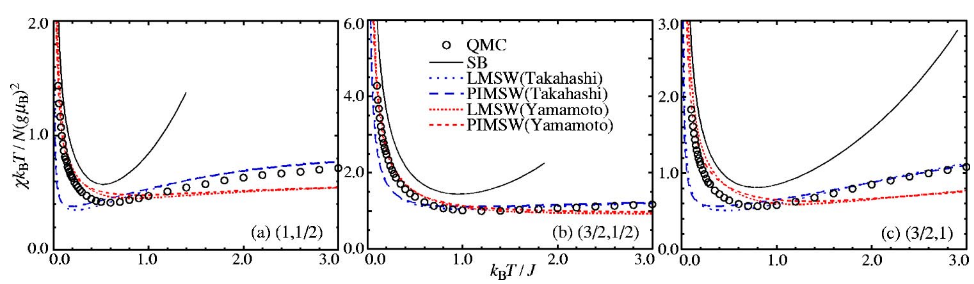

FIG. 4. (Color online) The Schwinger-boson (SB), linear-modified-spin-wave (LMSW), perturbational interacting-modified-spin-wave (PIMSW), and quantum Monte Carlo (QMC) calculations of the susceptibility-temperature product χT as a function of temperature for the spin-(S,s) ferrimagnetic Heisenberg chains. The modified spin waves are constructed in two different ways, the Takahashi scheme (Takahashi) and the different scheme (Yamamoto).

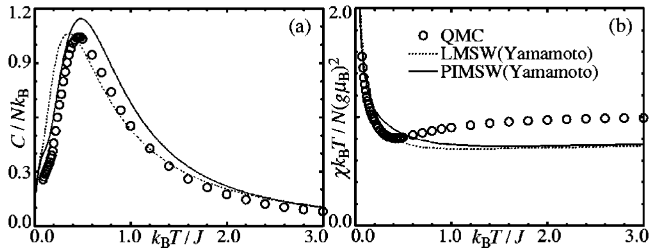

FIG. 5. The linear-modified-spin-wave (LMSW), perturbational interacting-modified-spin-wave (PIMSW), and quantum Monte Carlo (QMC) calculations of the specific heat C and the susceptibility-temperature product χT as functions of temperature for the spin-$\frac{1}{2}$ bond-tetrameric ferrimagnetic Heisenberg chain of $J_F = J_{AF}$. The modified spin waves are constructed on the different scheme.

paramagnetic behavior showing an antiferromagnetic increase. The modified spin waves much better describe the magnetic behavior than the Schwinger bosons. The spin waves modified along with the Takahashi scheme better work at high temperatures, while those along with the different scheme precisely reproduce the low-temperature behavior. Both calculations converge into the paramagnetic behavior $\chi k_B T/N(g\mu_B)^2 = [S(S+1)+s(s+1)]/3$ at high temperatures, whereas the Schwinger-boson mean-field theory again breaks down at the particular temperature (3.1). Considering that numerical tools less work at low temperatures, we realize the superiority of the different scheme-based modified spin-wave theory all the more.

Finally we calculate another type of ferrimagnet in order to demonstrate the constant applicability of the present different scheme. Figure 5 shows the thermodynamic properties of the ferromagnetic-ferromagnetic-antiferromagnetic-antiferromagnetic bond-tetrameric spin-$\frac{1}{2}$ Heisenberg chain,

$$\mathcal{H} = \sum_{n=1}^{N} [J_{AF}(\boldsymbol{S}_{4n-3}\cdot\boldsymbol{S}_{4n-2} + \boldsymbol{S}_{4n-2}\cdot\boldsymbol{S}_{4n-1}) - J_F(\boldsymbol{S}_{4n-1}\cdot\boldsymbol{S}_{4n} + \boldsymbol{S}_{4n}\cdot\boldsymbol{S}_{4n+1})], \tag{3.2}$$

where we have set all the g factors equal for simplicity. The different modified spin-wave scheme again successfully reproduces the Schottky peak of the specific heat. The interacting modified spin waves further interpret the low-temperature shoulderlike structure. The characteristic minimum of the susceptibility-temperature product is unfortunately less reproduced but the calculation again correctly gives the paramagnetic susceptibility at sufficiently high temperatures. A recent experiment[65] on a single-crystal sample of $Cu(C_5H_4NCl)_2(N_3)_2$,[7] which may be described by the Hamiltonian (3.2), has reported that the specific heat exhibits a double-peaked structure as a function of temperature. There is indeed a possibility of an additional peak appearing at low temperatures as the ratio J_F/J_{AF} moves away from unity.[66] However, no parameter assignment has yet succeeded in interpreting all the observations consistently. There are further chemical attempts to synthesize novel ferrimagnets. Organic ferrimagnets[6,67] are free from magnetic anisotropy and thus suitable for analyzing in terms of the modified spin waves.

IV. SUMMARY AND DISCUSSION

We have demonstrated the Schwinger-boson mean-field representation and the modified spin-wave treatment of one-dimensional Heisenberg ferrimagnets. The Schwinger bosons form an excellent language at low temperatures but rapidly lose their validity with increasing temperature. The modified spin-wave theory is more reliable in totality provided the number of bosons is controlled without modifying the native energy structure. On the other hand, the Schwinger-boson representation can be extended to anisotropic systems[68] more reasonably because it is rotationally invariant in contrast to the modified spin-wave theory. While the temperature dependence of the antiferromagnetic excitation gap $\omega_{k=0}^{+}$ is left to solve experimentally, we are now convinced that the bosonic languages remain effective in low dimensions and may be applied to extensive ferrimagnets.[69] Besides ground-state properties and thermodynamics, quantum spin dynamics[70,71] can be investigated through the modified spin-wave scheme.

We further mention our findings in the antiferromagnetic limit with the view of realizing the close relation between the two bosonic languages. We equalize s with S and set $2N$, the number of spins, equal to L for the Hamiltonian (2.1). At $S = s$, the ground-state sublattice magnetization (2.37) diverges and therefore the different modified spin-wave scheme is no more applicable. We have to settle the quantum, as well as thermal, divergence inevitably employing the

TABLE II. The Schwinger-boson (SB), linear-modified-spin-wave (LMSW), perturbational interacting-modified-spin-wave (PIMSW), full-diagonalization interacting-modified-spin-wave (FDIMSW), and quantum Monte Carlo (QMC)[72] calculations of the ground-state energy E_g and the lowest excitation gap Δ_0 for the spin-S antiferromagnetic Heisenberg chains (Ref. 72).

	$S=1$		$S=2$		$S=3$	
Approach	E_g/LJ	Δ_0/J	E_g/LJ	Δ_0/J	E_g/LJ	Δ_0/J
SB	−1.396148	0.08507	−4.759769	0.00684	−10.1231	0.00295
LMSW	−1.361879	0.07200	−4.726749	0.00626	−10.0901	0.00279
PIMSW	−1.394853	0.07853	−4.759760	0.00655	−10.1231	0.00287
FDIMSW	−1.394617	0.08507	−4.759759	0.00684	−10.1231	0.00295
QMC	−1.401481(4)	0.41048(6)	−4.761249(6)	0.08917(4)	−10.1239(1)	0.01002(3)

Takahashi scheme. Besides the perturbational treatment of $\mathcal{H}_0$, we may consider the full diagonalization of $\mathcal{H}_1+\mathcal{H}_0$, where the ground-state energy and the dispersion relations are still given by Eqs. (2.31) and (2.32), respectively, but with θ_k satisfying

$$\tilde{\gamma}_1(k)+\gamma_0(k)=0. \tag{4.1}$$

Such an idea applied to ferrimagnets ends in gapped ferromagnetic excitations and misreads the low-energy physics. The perturbational series-expansion approach is highly successful in the case of ferrimagnets.[32,33] Focussing our interest on Haldane-gap antiferromagnets, we list the bosonic calculations of the ground-state properties in Table II. The bosonic languages interpret the ground-state correlation very well but underestimate the Haldane gap considerably. Indeed they cannot detect the topological terms responsible for vanishing gap,[73] but they are still qualitatively consistent with the nonlinear-σ-model quantum field theory, yielding the low-temperature limiting behavior $\omega^{+}_{k=0}-\Delta_0\propto \mathrm{e}^{-\Delta_0/T}$ (Refs. 41, 61) and the large-spin asymptotic behavior $\Delta_0\propto \mathrm{e}^{-\pi S}$.[44,46,73] The Schwinger-boson mean-field theory and the full-diagonalization interacting modified spin-wave treatment give the same estimate of the Haldane gap. The Schwinger-boson dispersion relation (2.10) indeed coincides analytically with that of the full-diagonalization interacting modified spin waves at zero temperature. This is interesting but not so surprising, because the Holstein-Primakoff bosons (2.18) are obtained by replacing both $a_{n\uparrow}$ ($b_{n\downarrow}$) and $a^\dagger_{n\uparrow}$ ($b^\dagger_{n\downarrow}$) by $\sqrt{2S-a^\dagger_{n\downarrow}a_{n\downarrow}}$ ($\sqrt{2s-b^\dagger_{n\uparrow}b_{n\uparrow}}$) in the transformation (2.2).

Figure 6 shows the thermodynamic calculations for the spin-1 antiferromagnetic Heisenberg chain. We learn that the Schwinger-boson mean-field theory does not work at all for spin-gapped antiferromagnets at finite temperatures, which is in contrast with its fairly good representation of the low-temperature thermodynamics for ferrimagnetic chains. On the other hand, the modified spin-wave treatment maintains its validity to a certain extent. Indeed the Takahashi scheme still fails to reproduce the antiferromagnetic Schottky-type peak of the specific heat, but it describes the susceptibility very well except for the low-temperature findings attributable to the underestimate of the Haldane gap. We may expect the modified spin waves to efficiently depict the dynamic, as well as static, susceptibility for extensive spin-gapped antiferromagnets including spin ladders.[74] As for the thermal properties of one-dimensional antiferromagnets, whether spin gapped or not, there is a possibility of a fermionic language,[75,76] which is in principle compact, being superior to any bosonic representation.

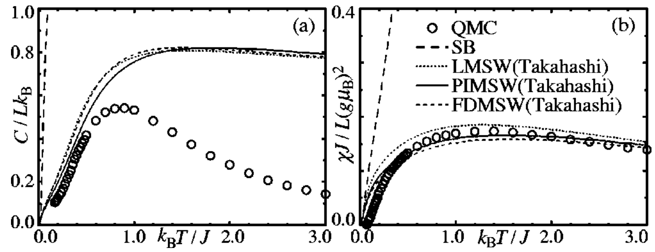

FIG. 6. The linear-modified-spin-wave (LMSW), perturbational interacting-modified-spin-wave (PIMSW), full-diagonalization interacting-modified-spin-wave (FDIMSW), and quantum Monte Carlo (QMC) calculations of the specific heat C and the susceptibility χ as functions of temperature for the spin-1 antiferromagnetic Heisenberg chain. The modified spin waves are constructed on the Takahashi scheme.

In the case of ferromagnets, the Holstein-Primakoff bosons are already diagonal in the momentum space,[37,56] suggesting no quantum fluctuation in the ground state, and therefore the present scheme turns out equivalent to the Takahashi scheme. The different-scheme-based modified spin-wave theory is the very method for low-dimensional ferrimagnets and is ready for extensive explorations.

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