Spin gaps—the energy gaps in magnetic excitation spectra—have been attracting much interest in recent years. Haldane [1,2] pioneeringly pointed out possible gapped excitations in one-dimensional Heisenberg antiferromagnets with integral spins. There followed intergap excitations in one-dimensional Heisenberg antiferromagnets [3]. Haldane [1,2] pioneeringly pointed out possible gapped excitations in one-dimensional Heisenberg antiferromagnets with integral spins. There followed intergap excitations in one-dimensional Heisenberg antiferromagnets [3].

The conventional spin-wave theory [19,20] applied to Heisenberg single-rung ladders ends in diverging sublattice magnetizations and vanishing gap [7]. We therefore employ the sublattice-symmetric spin-wave theory [21,22], originally applied to square-lattice antiferromagnets. Handling Holstein-Primakoff bosons, we make our first attempt to describe ladder antiferromagnets in terms of spin waves. We make interesting explorations into ladder systems featuring fermions versus bosons in an attempt to provide an intuitive picture for understanding the numerical findings and to reveal novel quantum phenomena peculiar to low dimensions.

We study the spin-$\frac{1}{2}$ antiferromagnetic Heisenberg model on two-leg ladders:

$$\mathcal{H} = J \sum_{i=1}^{2} \sum_{j=1}^{N} S_{i,j} \cdot S_{i,j+1} + J' \sum_{j=1}^{N} S_{1,j} \cdot S_{2,j},$$

which has isotropic nearest-neighbor exchange interactions along the chains (J) and along the rungs (J'). In the two extreme cases of $J' = 0$ and of $J = 0$, the low-energy properties are well known. The decoupled chains are critical, whereas the decoupled rung dimers are massive. The intermediate region with $J' \approx J$ is thus interesting, but most of model materials lie in the regime of large $J'$, such as Cu$_2$(C$_5$H$_{12}$N$_2$)$_2$Cl$_4$, Cu$_2$(C$_5$H$_{12}$N$_2$)$_2$Br$_4$ with $J' \approx 5J$ [23], Cu$_2$(C$_5$H$_{12}$N$_2$)$_2$Br$_4$ with $J' \approx 7J$ [24], and (C$_5$H$_{12}$N)$_2$CuBr$_4$ with $J' \approx 3.5J$ [25]. Hence much attention is paid to a copper oxide SrCuO$_3$, comprising two-leg ladders with $J' \approx J$ [26]. We consider the case of $J' = J$ unless otherwise noted. It is along a snake-like path [16,17], $(i,j) = (1,1) \to (2,1) \to (2,2) \to (1,3) \to \cdots$, that we align spinless fermions (SFs). When we introduce renumbered spin operators $\tilde{S}_{i,j} = S_{i,j}$ (SFs) for odd (even) $j$'s, where $\bar{i} = 3 - i$, the SFs are created as $c_{\bar{i},j}^\dagger = \tilde{S}_{\bar{i},j} \exp[-i\pi(\sum_{n=1}^{i-1} \tilde{S}_{m,n}^\dagger \tilde{S}_{m,n} + \sum_{m=1}^{i-1} \tilde{S}_{m,j} \tilde{S}_{m,j})]$. The fermionic Hamiltonian is given by

$$\mathcal{H} = \frac{1}{2} \sum_{i=1}^{2} \sum_{j=1}^{N} \left[ J \left( c_{i,j}^\dagger c_{i,j+1} + c_{i,j+1}^\dagger c_{i,j} - c_{i,j}^\dagger c_{i,j} + \frac{1}{4} \right) + J' \left( c_{i,j}^\dagger c_{i,j+1} e^{-i\pi d_i} \left( c_{i,j+1}^\dagger c_{i,j+1} + c_{i,j}^\dagger c_{i,j} \right) + H.c. \right) + J' \frac{1}{2} \left( c_{i,j}^\dagger c_{i,j}^\dagger c_{i,j} + c_{i,j}^\dagger c_{i,j}^\dagger c_{i,j}^\dagger + 1 \frac{1}{4} \right) \right].$$

and we search for its mean-field solutions. Setting the...
thermal average $\langle c_{1,j}^\dagger c_{i,j} \rangle$ equal to $1/2$ under zero magnetization and defining a unitary transformation

$$\left( \frac{a_{1,k}}{a_{2,k}} \right) = \frac{1}{\sqrt{2|\varepsilon_k|}} \left( |\varepsilon_k| e^{\frac{i\pi}{2}} - |\varepsilon_k| \right) \left( c_{1,k} \right),$$

we can diagonalize the Hamiltonian as $\sum_k |\varepsilon_k| (|\alpha_{1,k} a_{1,k} - a_{2,k}^\dagger a_{2,k}|)$, where

$$\varepsilon_k = \left( \frac{1}{2} - \chi_0 \right) J' + \left( \frac{1}{2} - \chi_1 - 2|\chi_2|^2 - \chi_2^* \right) J \cos k$$

$$+ i \left( \frac{1}{2} - \chi_1 + 2|\chi_2|^2 + \chi_2^* \right) J \sin k,$$

with $\chi_0 = \langle c_{2,j}^\dagger c_{1,j} \rangle$, $\chi_1 = \langle c_{2,j}^\dagger c_{1,j+1} \rangle$, and $\chi_2 = \langle c_{1,j}^\dagger c_{2,j+1} \rangle$ to be self-consistently determined at each temperature in an approximation of the Hartree-Fock type, while within the Hartree-level approximation, $\varepsilon_k = J'/2 + i J \sin k$, suggesting a spin gap $J'/2$.

A bosonic approach starts from the Holstein-Primakoff transformation: $S_{i,j} = (2S - a_{1,j}^\dagger a_{1,j})^{1/2} a_{1,j}$, $S_{i,j}^2 = S - a_{1,j}^\dagger a_{1,j}$ for even $i+j$'s, while $S_{i,j} = a_{2,j}^\dagger (2S - a_{2,j}^\dagger a_{2,j})^{1/2}$, $S_{i,j} = -S + a_{2,j}^\dagger a_{2,j}$ for odd $i+j$'s. Then the Hamiltonian is expanded as $\mathcal{H} = \sum_{i=-2}^{\infty} \mathcal{H}_i$, where $\mathcal{H}_i$ is the $O(1/S^i)$ term and is taken into consideration up to $i = 0$. $\mathcal{H}_{-1}$ and $\mathcal{H}_0$ contain bilinear and biquadratic terms with respect to the bosonic operators and describe the linear and interacting spin waves, respectively [27]. In order to avoid quantum as well as thermal divergence of the sublattice magnetizations, the Bogoliubov transformation is carried out subject to the constraint that the sublattice magnetizations be zero [21,22]:

$$\sum_j a_{1,j}^\dagger a_{1,j} = \sum_j a_{2,j}^\dagger a_{2,j} = NS.$$

Compared with the conventional spin-wave theory, where spins on one sublattice point predominantly up, while those on the other predominantly down, the modified spin waves (MSWs) restore the sublattice symmetry. If we define the Bogoliubov transformation as

$$\left( \frac{a_{1,k}}{a_{2,k}} \right) = \frac{1}{\sqrt{2|\varepsilon_k|}} \left( \frac{x_{1,k}}{x_{2,k}} \right) \left( \frac{\beta_{1,k}}{\beta_{2,k}} \right),$$

$\mathcal{H}_{-1}$ is diagonalized with

$$x_{1,k} = \sqrt{\left( \frac{J' + 2J S + 2J \Delta}{\varepsilon_k} \right) + 1},$$

$$x_{2,k} = \sqrt{\left( \frac{J' + 2J S + 2J \Delta}{\varepsilon_k} \right) - 1 \operatorname{sgn}(J' - 2J \cos k)},$$

where $\lambda$ is the Lagrange multiplier due to the constraint (5) and $\varepsilon_k$ is the MSW dispersion relation:

$$\varepsilon_k = \sqrt{(J' + 2J S + 2J \Delta)^2 - [(J' - 2J \cos k) S]^2}.$$

The optimum thermal distribution functions of the MSWs are given by $\langle \beta_{i,k}^\dagger \beta_{i,k} \rangle \equiv \bar{n}_{i,k} = \langle e^{\varepsilon_k - \gamma S J \cos k} \rangle / \langle e^{\varepsilon_k} \rangle$. The spin gap by the linear MSWs (LMSWs), the interacting spin waves (IMSWs), the Hartree-level SFs (HSFs), and the Hartree-Fock-level SFs (HFSFs), and in Fig. 1 show them all together with numerical (Exact) findings [7] and strong-coupling-expansion (SCE) results [30]. $E_k/NJ = -3r/4 - 3/8r + O(r^{-2})$ and $\Delta/J = r - 1 + 1/2r + O(r^{-2})$, where $r = J'/J$. Unless $J'$ is sufficiently large, the ground-state energy is better described by the MSWs, while the spin gap by the SFs. When we focus on the interesting point $J' = J$, the IMSWs give the best estimate of the energy as $E_k/NJ = -1.144$, compared with the numerically exact value $-1.156$, whereas the HSFs give that of the gap as $\Delta/J = 1/2$, compared with 0.50(1). The HSF description is no more precise once the system moves away from this point, but the HFSF description remains quantitative in a wider range of $J'/J$. The MSWs are trivially less relevant to decoupled clusters, where no “wave” can spread over the system. The dispersion relation is also shown in Fig. 1. The band bottom is well reproduced by the SFs, while the band width by the MSWs. Since the MSWs considerably underestimate the spin gap, any thermally activated behavior at low temperatures should be observed through the SFs.

Secondly we calculate the thermal properties and compare them with quantum transfer-matrix (QTM) calculations [31]. The specific heat $C$ is shown in Fig. 2(a). The SFs well reproduce the Schottky peak, while the MSWs lose their validity with increasing temperature. The HSFs (HFSFs) underestimate the high-temperature

\[\text{FIG. 1. The ground-state energy (a) and the spin gap (b) as functions of } J'/J, \text{ and the dispersion relation of spin-triplet excitations (c).}\]
behavior \( C/Nk_B = (3k_BT/4J)^2 + O[(k_BT/J)^4] \) by a factor 2/3 (4/9) but complement numerical tools very well at low temperatures. If we approximate the dispersion relation of the low-lying excitations as

\[
E_n = \Delta + J\alpha(k - \pi)^2,
\]

where \( \Delta = J'/2 \) and \( a = J'/J \) in the Hartree approximation, while \( \Delta = [(1/2 - \chi_0)J' - (1/2 - \chi_1 - 2\chi_2^2 - \chi_2)J] \) and \( a = (1/2 - \chi_0)/(1/2 - \chi_1 - 2\chi_2^2 - \chi_2)J' + 4(\chi_2 + 2\chi_2^2)J/2\Delta \) in that of the Hartree-Fock type, the SFs illuminate the low-lying excitations as functions of temperature.

The MSWs are good at reproducing the overall behavior. All the calculations converge into the paramagnetic susceptibility \( 2S(S + 1)/3k_BT \) at high temperatures.

Lastly we investigate the nuclear spin-lattice relaxation rate in terms of the SFs, which are much better than the MSWs at describing the spin gap and therefore the low-temperature properties. Considering the electronic-nuclear energy-conservation requirement, the Raman process plays a leading role in the relaxation, which is formulated as

\[
\frac{1}{T_1} = \frac{4\pi\hbar(g\mu_B\gamma_N)^2}{3k_BT} \sum_n e^{-E_n/k_BT} \sum_{n,m} e^{-E_m/k_BT} \delta(E_n - E_m - \hbar\omega_N),
\]

where \( A \) is the hyperfine coupling constant between the nuclear and electronic spins, \( \omega_N \equiv \gamma_NH \) is the Larmor frequency of the nuclei with \( \gamma_N \) being the gyromagnetic ratio, and the summation \( \sum_n \) is taken over all the electronic eigenstates \( |n\rangle \) with energy \( E_n \). Taking account of the significant difference between the electronic and nuclear energy scales \( \langle \hbar\omega_N \rangle \lesssim 10^{-5}J \) and assuming a reasonable temperature range \( k_BT \lesssim \Delta \) for \( \text{SrCu}_2\text{O}_3 \) with \( \Delta/k_B \sim 420 \text{ K} \) [26], the relaxation rate is represented as

\[
\frac{1}{T_1} \sim \frac{(g\mu_B\hbar\gamma_N)^2}{4\pi\hbar J A} \int_0^{2\pi} \sum_{i,k} n_{i,k}(1 - \bar{n}_{i,k}) \frac{\hbar\omega_N}{(\hbar\pi)^2 + (\hbar\omega_N/J)^2} dk,
\]

where we have again employed the approximate dispersion (9). The term \( n_{i,k}(1 - \bar{n}_{i,k}) \) is the consequence of the principle of detailed balancing in a fermionic ensemble. At moderate fields and temperatures, \( k_BT \ll \Delta - g\mu_BH \), Eq. (13) can be further calculated analytically as

\[
\frac{1}{T_1} \sim \frac{(g\mu_B\hbar\gamma_N)^2}{2\pi\hbar JA} e^{-\Delta/k_BTcosh(g\mu_BH/k_BT)K_0\left(\frac{\hbar\omega_N}{2k_BT}\right)},
\]

where \( K_0 \) is the modified Bessel function of the second kind and behaves as \( K_0(x) \approx 0.11593 - \ln x \) for \( 0 < x \ll 1 \). Equation (13), together with its approximate expression (14), is plotted in Fig. 3.

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 was pointed out in a sophisticated numerical work [31] as well, may be observed at sufficiently low temperatures but is not yet verified for ladder antiferromagnets such as \( \text{SrCu}_2\text{O}_3 \) owing to the magnetic impurities masking the intrinsic properties. Equation (13) deviates from the simple spin-gap-activated behavior (14) for \( k_BT > 0.2J \approx 0.4\Delta \), which is interestingly consistent with the criterion for the breakdown of the simple two-magnon scheme observed in a spin-1 Haldane-gap antiferromagnet \( \text{AgVP}_2\text{S}_6 \) [32]. In
such a high-temperature range, we have to fully incorporate particle collisions into the calculation in order to describe the crossover to diffusive behavior, where an enhanced activation gap may be observed due to the temperature-dependent diffusion constant [33]. In the low-temperature range \( k_B T \lesssim 0.2J \), on the other hand, the relaxation rate should strictly follow the expression (14): *With increasing field, \( 1/T_1 \) first decreases logarithmically and then increases exponentially.* 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 ferro- and ferrimagnets may exhibit similar field dependence [34]. The present logarithmic field dependence at low temperatures should and could be distinguished from the \( 1/\sqrt{H} \) or \( \ln(1/H) \) dependence of diffusion-dominated dynamics [35,36] at high temperatures. The following exponential increase originates in the spin-1 excited state lowering in energy with increasing field. If we consider the strong-correlation compound SrCu2O3 with \( J/K_A \approx 840 \text{ K} [26] \), the minimum is supposed to appear at \( H \approx 20 \text{ T} \). In the case of molecular-based ladder systems [23–25] with much smaller exchange interactions, \( 1/T_1 \) should reach a minimum at much smaller fields. We expect that such intrinsic features of the low-energy dynamics of ladder antiferromagnets will be found experimentally.

We have demonstrated new schemes of investigating Heisenberg two-leg ladder antiferromagnets. Although both fermionic and bosonic approaches possess advantages of their own, we stress the SFs along the snake-like path as one of the best languages for the present system. One of the fatal weak points in the MSW description is nonvanishing specific heat at high temperatures. The endlessly increasing energy with increasing temperature is because of the temperature-dependent energy spectrum (8), where \( \Lambda \), playing the role of the chemical potential, turns out a monotonically increasing function of temperature. Such a difficulty can be overcome in ferromagnets exhibiting noncompensating sublattice magnetizations [37,38] but plagues bosonic approaches applied to antiferromagnets. The SFs well reproduce the spin gap and thus reliably describe the low-temperature properties. *One of the most interesting findings is the novel field dependence of \( 1/T_1 \).* Relevant relaxation-time measurements are strongly encouraged.

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