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Excitation spectra of the linear alternating antiferromagnet

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The linear, spin-$\frac{1}{2}$, alternating Heisenberg chain has attracted theoretical and experimental attention from physical chemists for about two decades, particularly in relation to spin exciton theory and the properties of linear, exchange-coupled free radicals. The model is somewhat unfamiliar to physicists but has become of increasing interest recently, primarily because of its relation to spin-Peierls transition systems. A striking feature of this model is that it has so far proved resistant to any form of analytic attack. Existing theories are therefore all approximate, and are not in agreement with one another. In particular, there is disagreement about the existence of an energy gap in the excitation spectrum for nonzero alternation, such a gap being crucial to spin-Peierls theory and spin exciton theory. In this paper we employ the method which has so far proved more reliable than any other approximate technique, namely the method of extrapolating exact finite-chain calculations to the thermodynamic limit. Our study is an extension of earlier work in this direction, and focuses on the nature of the ground state and on low-lying excitations in general, and the existence and properties of the gap in particular. We introduce the features of the linear alternating antiferromagnet through an initial description of the spin-Peierls transition and with brief reference to organic free radicals and spin exciton theory. This is followed by a survey of existing approximate theories. Features of the excitation spectrum are discussed and finite-chain extrapolations for the ground-state energy and energy gap as a function of alternation are presented. Comparisons are made with similar procedures performed on exactly solvable models, as a test of the expected accuracy of the extrapolations. Excitation spectra for a variety of other alternating models, classical and quantum, are calculated and surveyed comparatively. An unusual variety of behavior is observed, with striking differences between quantum and classical systems. Finally, a detailed comparison is made between our results and those of other approximate methods, including the new quantum renormalization-group approach. Particular attention is paid to values for the $T=0$ spin-Peierls critical exponents.

I. INTRODUCTION

A spin-Peierls transition occurs when a system of uniform, isotropic, Heisenberg antiferromagnetic linear chains undergoes a transformation to a system of dimerized or alternating antiferromagnetic linear chains. This transition may be regarded as the insulating analog of the familiar Peierls transition in a one-dimensional conductor, although in the spin-Peierls case there is, of course, no significant change in the electrical conductivity as the temperature is lowered through the transition temperature T_{SP} (an insulator-to-insulator rather than a metal-to-insulator transistor). Strictly both Peierls and spin-Peierls transitions arise through the effects of weak interchain coupling; interchain spin-phonon coupling occurs in the spin-Peierls case.[1] Spin-Peierls transitions thus belong to the general class of magnetoelastic transitions. It must be supposed that in spin-Peierls systems the underlying crystal lattice is "soft," i.e., subject to distortion. For a more rigid lattice one might expect in-

stead the occurrence of the familiar quasi-one-dimensional (quasi-1D, i.e., 3D) magnetic ordering.

At a spin-Peierls transition the lattice distorts in such a way that successive spin-bearing ionic complexes move alternately closer and further apart. Instead of a single exchange constant J for the uniform chain, there are now two alternating exchange constants J_1 and J_2, with $J_1 > J_2$, for example. A simple description of this dimerization effect implies that $J_1 = J(1+\delta)$ and $J_2 = J(1-\delta)$, where δ is an interaction modulation parameter resulting from a shift in interionic distance. This implies the constraint $J_1 + J_2 = 2J$.

The Hamiltonian for a linear, alternating magnetic chain of an even number of spins (i.e., the magnetic part of the Hamiltonian for a spin-Peierls system) may be written

$$H = -2J_1 \sum_{i=1}^{N/2} \vec{S}_{2i-1} \cdot \vec{S}_{2i} - 2J_2 \sum_{i=1}^{N/2} \vec{S}_{2i} \cdot \vec{S}_{2i+1} , \tag{1.1}$$

where for much of this discussion, $J_1, J_2 < 0$, i.e., the systems are antiferromagnetic; we also assume N even. It is convenient to define an alternation parameter $\alpha = J_2/J_1$, and the Eq. (1.1) appears as[2]

$$H = -2J_1 \sum_{i=1}^{N/2} (\vec{S}_{2i-1} \cdot \vec{S}_{2i} + \alpha \vec{S}_2 \cdot \vec{S}_{2i+1}) . \tag{1.2}$$

Although theoretical descriptions of the spin-Peierls dimerization phenomenon have been available for almost two decades, experimental data which exhibit with classic simplicity the features expected of a spin-Peierls transition, have been observed only recently.[3,4] The dimerization process has been confirmed by x-ray studies.[5] The static low-field (zero-field) susceptibility data closely resemble the schematic curve shown in Fig. 1. For $T > T_{SP}$, the data show a very good fit to the theoretical curve for a uniform linear, spin-$\frac{1}{2}$, antiferromagnet.[6] Below T_{SP} the experimental data, instead of approaching a nonzero value as $T \to 0$ following the theoretical curve (now shown dashed) show a sharp, apparently exponential, drop to zero. The striking feature that the $T < T_{SP}$ data are independent of single-crystal orientation indicates that the spin-isotropic, or Heisenberg, character of the associated magnetic system is preserved below the transition. The essentially exponential character of the susceptibility below T_{SP} indicates that an energy gap has opened up between the nonmagnetic, total-spin $S = 0$, singlet ground state and the (band of) lowest-excited $S = 1$ triplet states. The situation is sketched in Fig. 2.[7] In Fig. 2(a) the lowest-lying excitations for a uniform linear Heisenberg antiferromagnet are shown. The excitation branches follow the well-known $|\sin k|$ dispersion law calculated analytically by des Cloizeaux and Pearson.[8] It is now known that these states actually form the lower edge of a two-parameter continuum of degenerate singlet and triplet delocalized (spin-wave) excitations.[9,10] Figure 2(b) shows schematically the effect on the excitation spectrum of introducing alternation. Since the unit cell is now double, the Brillouin zone is halved, extending from $\pi/2$ to $-\pi/2$ instead of from π to $-\pi$. More importantly, the excitation branches are now detached from the singlet ground state by a minimum excitation gap ΔE. The sketch in Fig. 2(b) is based on various approximate theories, such as that of Bulaevskii[11] and others.[12–14] The sketch is also in accordance with numerical studies of Duffy and Barr for a relatively large degree of alternation.[15]

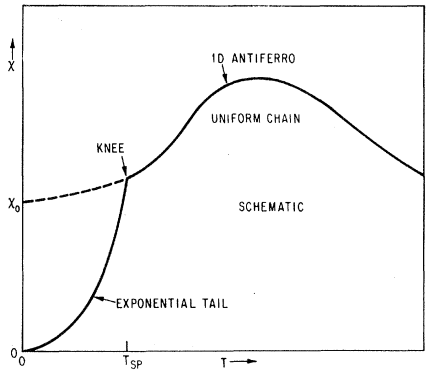

FIG. 1. Characteristic zero-field susceptibility, χ vs T, for a system showing a spin-Peierls transition. Above the transition the system behaves as an assembly of uniform Heisenberg antiferromagnetic chains. Below the transition, T_{SP}, χ drops sharply, going exponentially to zero, whereas the uniform curve, now shown dashed, continues on to a nonzero value, χ_0.

The field of physical chemistry provides a second area of application for linear, antiferromagnetic spin-$\frac{1}{2}$ Heisenberg magnets. Organic molecular crystals exist containing linear chains of exchange-coupled free radicals. Since the free-radical solids are insulators or semiconductors containing tightly bound electrons, the exchange problem along each chain reduces to that of the alter-

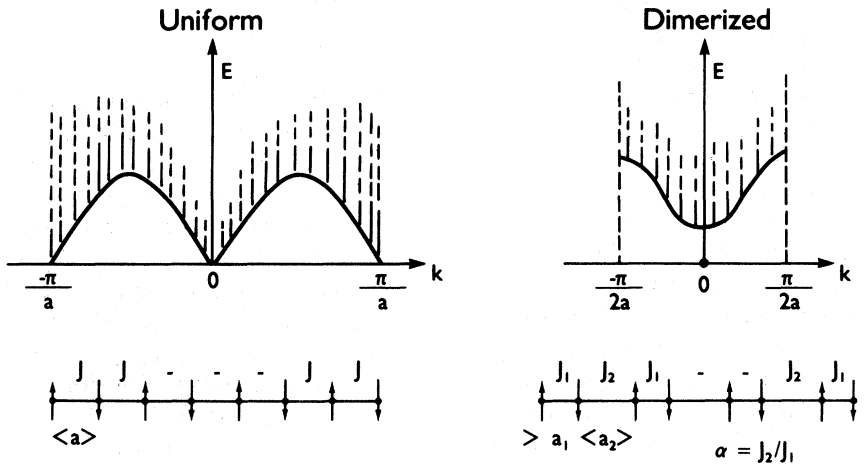

FIG. 2. (a) Schematic excitation dispersion spectrum for a uniform Heisenberg antiferromagnetic chain. (b) Schematic excitation dispersion spectrum for an alternating (dimerized) Heisenberg antiferromagnetic chain. Note the energy gap at $k=0$ and the halving of the Brillouin zone.

nating linear Heisenberg antiferromagnet [Eqs. (1.1) or (1.2)]. Spin-exciton theory describes the magnetic excitations of such crystals, and is based on the picture of a singlet ground state with a band of triplet excitations above an excitation gap [as in Fig. 2(b)]. It has further been suggested that close to the uniform limit, the triplet spin excitons are delocalized in character (Wannier spin excitons), whereas close to the fully alternating (noninteracting-dimer) limit, the excitons have a bound character (Frenkel spin excitons).[16]

It is clear from the above discussion that the existence of an energy gap in the alternating, antiferromagnetic Heisenberg chain for all degrees of alternation is fundamental to both spin-Peierls theory and spin-exciton theory. (In addition, a precise knowledge of the character of the low-lying spectral excitations would determine the validity of spin-exciton theory.) Unfortunately, the general problem of the spin-$\frac{1}{2}$ alternating linear chain appears analytically intractable at this time.[17] It is not even feasible, apparently, to attain the limited objective of using rigorous methods of proof to demonstrate the existence of a gap. Nevertheless, attention has been called to the general question of the low-lying spectral excitations of alternating chains; in particular, the existence of a gap.

A. Existing theories

Let us first review the status of theories which predict a gap and then review theories and arguments which do not. An important approximate calculation on which much subsequent work has rested[18] is the Bulaevskii Hartree-Fock approach.[11] The corresponding theory for the uniform limit[19] has been surprisingly successful in correctly predicting the general qualitative features of the uniform antiferromagnet where other approximate methods have failed rather badly, as discussed in Refs. 20 and 21. Subsequently numerical calculations by Duffy and Barr[15] in the spirit of earlier work by Bonner and Fisher[6] were undertaken to provide results more quantitative than those of the Bulaevskii approach for comparison with experimental data on various organic free radicals. Specifically, the exact eigenvalues and thermodynamic properties of the alternating-spin Hamiltonian for short spin-$\frac{1}{2}$ rings of $N=4$, 6, 8, and 10 spins were calculated for alternation values $\alpha=0.2$, 0.4, 0.6, and 0.8. Rapid convergence of the finite-N alternation energy gap, i.e., the gap between the singlet ground state and the first excited (triplet) state(s), enabled Duffy and Barr to conclude that it was most unlikely that the gap would vanish in the limit $N\rightarrow\infty$ for $\alpha\lesssim 0.6$. The calculations which are the focus of this paper are, in fact, an extension of the Duffy-Barr approach to rings of length $N=12$, together with a detailed examination of the character of the low-lying excited states and a variety of extrapolation techniques to determine the large-N behavior of the alternation energy gap for all α.

Other many-body theories applied to this problem include the quasiboson calculation of Montgomery,[12] which gives a gap for all $\alpha<1$, vanishing in the limit $\alpha=1$. Unfortunately, the theory is

in disagreement with the rapidly converging Duffy-Barr calculations near the dimer limit, and must be considered less reliable than the Bulaevskii theory.[11] Other theories[13,14] predict a gap for all α, including the uniform limit, where a gap is known rigorously not to exist, and therefore cannot be considered reliable.

A relatively high-order (third-order) perturbation-theory calculation by Harris,[17] where the interdimer or weak interpair interaction is treated as a perturbation on a noninteracting pair model, yields an expansion in α whose convergence indicates strongly that the energy gap does not vanish for $\alpha \lesssim 0.6$, in accordance with the finite-cluster calculations of Duffy and Barr. A nonvanishing energy gap for $\alpha < 0.6$ is in accordance with extensive experimental measurements on copper nitrate, whose dominant underlying magnetic structure is argued to correspond to an alternating, Heisenberg antiferromagnetic chain with an alternation parameter $\alpha \simeq 0.3$.[22,23,24] Recently, calculations have been performed on a Heisenberg, spin-$\frac{1}{2}$, alternating, antiferromagnetic linear chain in a continuum approximation rather than on a discrete lattice. The theoretical approach is an extension of a calculation by Luther and Peschel[25] on the corresponding uniform continuum model. This recent work of Cross and Fisher[26] predicts the existence of a gap for all $\alpha < 1$, but the effects of the continuum approximation are hard to assess.

Arguments which have raised doubts concerning the existence of a singlet-triplet gap in the linear, alternating antiferromagnetic chain are based on spin-wave arguments and also the isotropic spin character of the Heisenberg Hamiltonian. Classical spin-wave discussions (equivalent to spin infinity) led Ginzberg and Fain[27] to conclude that the alternating Heisenberg chain possesses only an acoustic (i.e., gapless) branch in its excitation spectrum. Further studies of classical, spin-∞ linear chains will be presented later in this paper. The isotropic character of the spin Hamiltonian is related to the interesting question of Goldstone modes.[28]

In magnetic terms, a system with isotropic spin symmetry is expected to show a gapless "acoustic" excitation branch emanating from the ground state. This situation actually occurs in the case of the uniform Heisenberg ferromagnet and antiferromagnet, and in the case of the alternating Heisenberg ferromagnet for all α. However, the Goldstone theorem depends for its validity on the presence of an infinitely degenerate ground state. For the case of the spin-$\frac{1}{2}$, alternating ferromagnet, the ground state is $(N+1)$-fold degenerate for $\alpha > 0$ (including the uniform limit). Hence, there is no gap. In the case of the alternating antiferromagnet, however, the ground state is known to be a (nondegenerate) singlet,[29] in which case the Goldstone theorem need not necessarily apply. Nevertheless, the stimulus was present to examine the validity of the Bulaevskii approximation, which predicts a gap for $\alpha < 1$. The decoupling procedures in the Bulaevskii theory fail to preserve the spin-rotational symmetry associated with a gapless mode. Therefore Drawid and Halley (DH) formulated a Green's-function approximation which preserved spin-rotational invariance, and the outcome is that two branches always appear; a gapless acoustic, and an optic branch with a gap. (Note that in this paper we use the adjective "acoustic" for dispersion branches without a gap, and the adjective "optic" for branches with a gap, without consideration of the nature of the excitations corresponding to the particular branches.) Figure 3 is a sketch of the DH predictions, based on Fig. 2 of their paper.[30] They give a clear discussion of the significance of their results in comparison with other predictions then available, and this is summarized in Fig. 4, based on Fig. 1 of their paper, which is a qualitative sketch of gap ΔE as a function of alternation parameter α. The possibilities include curves of types *a*, *b*, *c*, and *d*. The DH approach gives curve *a*. No gap is present for all $\alpha > 0$. At $\alpha = 0$, corresponding to an assembly of noninteracting spin dimers, a gap discontinuously appears. Our feeling is that curve *a* is highly implausible in view of the

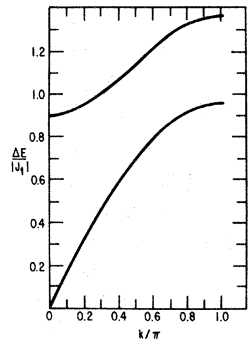

FIG. 3. Schematic predictions of an approximate approach to the spin-$\frac{1}{2}$, alternating Heisenberg antiferromagnetic chain (based on Fig. 2 of Ref. 30). A gapless acoustic mode is predicted in addition to an optic mode with a gap.

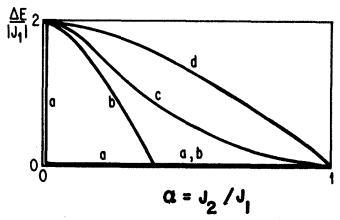

FIG. 4. Qualitative sketch of gap ΔE as function of alternation parameter α (based on Fig. 3 of Ref. 30). Curve a implies no gap for all $\alpha > 0$. For curve b the gap vanishes midway through the range of alternation. Curves c and d show energy gaps persisting to the uniform limit, i.e., for all $\alpha < 1$.

rapid convergence of finite-N chains first observed by Duffy and Barr[15] in the region close to the dimer limit (i.e., $\alpha \lesssim 0.6$), and also the rapid convergence of the perturbation series of Harris[17] in the same alternation region. Curve b, where the gap disappears midway through the region $0 \leq \alpha \leq 1$, is an interesting possibility not inconsistent with the Duffy-Barr and Harris calculations. Such a possibility would, however, be in conflict with the smoothness-universality hypothesis which states, in essence, that no drastic change in the critical behavior of a system is expected as a linear parameter in the Hamiltonian is varied, unless a fundamental change in the symmetry character of the Hamiltonian occurs.

Our intuition, therefore, lies in favor of excitation curves of type c or d in Fig. 4, where the gap disappears only in the uniform ($\alpha = 1$) limit. Extensive numerical calculations are in agreement with our intuition and, further, suggest a gap curve of type d rather than type c. A similar type of curve results from the recent Cross and Fisher continuum linear-chain calculation.[26] Of course, numerical calculations for small finite systems cannot "prove" the existence or otherwise of a spectral energy gap for an infinite system. However, such numerical extrapolations have proved reliable in the past, as will be discussed later in the paper, and we feel the weight of evidence for the presence of a gap for all $\alpha < 1$ is now very strong. This conclusion is reinforced by very recent and ongoing zero-temperature renormalization-group (RG) calculations[31–33] which reveal the existence of two fixed points for the alternating, antiferromagnetic Heisenberg chain. One fixed point occurs at $\alpha = 0$ (dimer limit) and is stable; the other occurs at $\alpha = 1$ (uniform limit) and is unstable. Therefore systems with initial values $\alpha < 1$ must flow (with increasing RG iteration) into the stable fixed point $\alpha = 0$. Therefore, for $\alpha < 1$, the system is equivalent to a completely dimerized system as far as its low-lying energy-state symmetries are concerned.

The organization of this paper is as follows. In Sec. II the character of the low-lying spectral excitations, including the ground state, deduced from exact finite-chain calculations is investigated. The results of numerical extrapolations are presented in Sec. III. Details of the quantum-mechanical calculations are given in Appendix A and details of the various extrapolation techniques in Appendix B. In Sec. IV interesting exact calculations on various solvable alternating-spin chains are summarized and their significance discussed. Section V gives a critical discussion of the significance of the various results.

II. FEATURES OF THE EXCITATION SPECTRUM

To gain an understanding of the nature of the low-lying excitations of finite, antiferromagnetic chains, it is useful to being with an examination of the dispersion spectrum for a *uniform* ring of $N = 12$ spins. The dominant, low-lying states are shown in Fig. 5, plotted in terms of excitation energies from the nondegenerate, singlet, antiferromagnetic ground state, as a function of wave vector k. The states are characterized in terms of multiplicity, i.e., singlet, triplet, quintet, etc., and

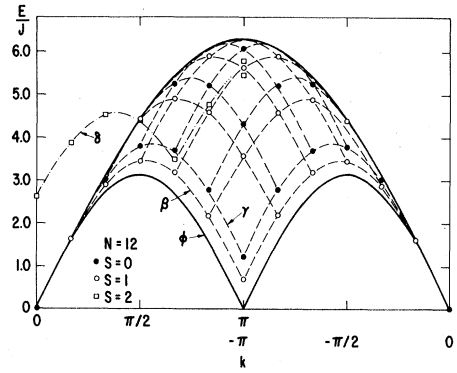

FIG. 5. An exact calculation of the lowest-lying excitations of a finite, uniform Heisenberg antiferromagnetic ring with $N = 12$. Singlet, triplet, and quintet states only are featured. The nature of the various dispersion branches β, γ, δ, and ϕ is explained in the text.

are arranged in sets of dispersion branches. The assignment prescription for the states is an extension of the Yang picture of hole-particle excitations from a filled Fermi sea. In Fig. 5 we also show, for comparative purposes, branch ϕ, which is the lowest-lying excitation branch for $N\to\infty$. Branch ϕ has the des Cloizeaux–Pearson[8] dispersion relation

$$E_1(k)=\pi J\,|\sin k|\ . \tag{2.1}$$

For $N=12$ the most important excitation branch is branch β containing only triplet states. As $N\to\infty$, branch β approaches the limiting dispersion value (2.1) as $1/N$. However, these states are not the only single particle-hole excitations. There are altogether a total of $N(N+2)/8$ low-lying triplets, whose relation to the β states is shown by the dashed dispersion curves in Fig. 5, and which form a spin-wave double continuum as $N\to\infty$ (two-parameter continuum) whose lower limit is the des Cloizeaux–Pearson dispersion curve (2.1) and whose upper limit is the envelope dispersion curve,[34,8,10,35]

$$E_2(k)=2\pi\,|\sin k/2|\ , \tag{2.2}$$

which clearly has twice the amplitude and half the periodicity of (2.1). However, an interesting feature of Fig. 5 is the existence of a second double continuum of singlet states, each of which "shadows" a corresponding triplet state. In the limit $N\to\infty$, numerical calculations show that the singlet states become degenerate with the triplet states. This numerical observation is in agreement with a prediction of Johnson *et al.*[36,37] that the low-lying spectral excitations should have a fourfold degeneracy. This two-parameter singlet continuum arises out of dispersion branch γ. In addition to the singlet and triplet excitations, low-lying quintet states appear, resulting from two-particle, two-hole excitations in the Yang prescriptions (see dispersion branch δ). However, the states of importance for this study are the ground state, the triplet states of dispersion branch β, and the singlet states of dispersion branch γ. While one is not sure, from studies on finite rings, that higher-order excitations may not become important in the limit $N\to\infty$, it is unlikely that we are neglecting any relevant states. This belief is supported by comparable finite-ring studies in spin-$\frac{1}{2}$ XY systems, where the behavior in the $N\to\infty$ limit is known exactly.

In Fig. 6, which also refers to the uniform limit, we see again the triplet branch β of Fig. 5. It

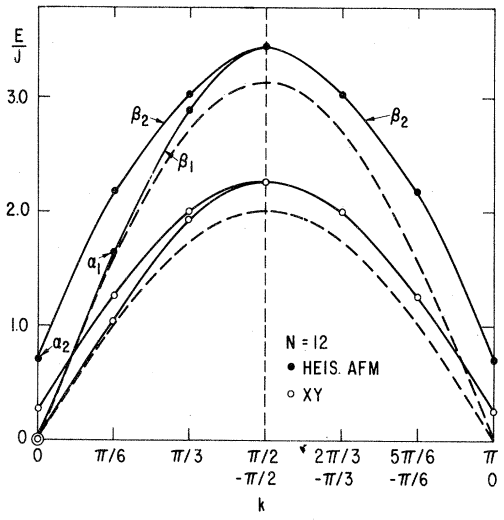

FIG. 6. A comparison of corresponding triplet dispersion branches for the $N=12$ uniform Heisenberg model and the $N=12$ XY model. The significance of the two branches β_1 and β_2 and the states α_1 and α_2 is explained in the text. The dashed curves show the exact $N\to\infty$ results for each model.

should be noted that the β branch for small finite N is not symmetric about $k=\pi/2$, as in the $N=\infty$-limiting dispersion curve of des Cloizeaux–Pearson. Figure 6 shows that a similar phenomenon occurs in the case of the XY model. It appears that there is a breakdown of symmetry for finite N which is restored in the $N\to\infty$ limit. In particular, the state at $k=\pi$ is split off from the ground state by an energy gap vanishing as $1/N$. To simulate the situation when alternation is present, part of branch β is reflected about $k=\pi/2$ to yield a reduced Brillouin zone of 0 to $\pi/2$ (strictly $-\pi/2$ to $\pi/2$). The two branches in the reduced zone are denoted β_1 and β_2. In considering the possibility of a Drawid-Halley (DH) (Ref. 30) excitation scheme one might suppose that the branches β_1 and β_2 remain nondegenerate in the alternating case in the limit $N\to\infty$, and, further, that the branch β_1 would remain attached to the ground state, becoming the DH acoustic branch and that the branch β_2 would become the optic branch, split off from β_1 and from the ground state by an optical excitation energy gap. However, our numerical extrapolations show rather conclusively that this is not so. It appears that both branches, β_1 and β_2, again become degenerate in the $N\to\infty$ limit, as in the case of the uniform chain, and that an excitation energy gap opens up

between the ground state and the lower end of the dispersion branches for all $\alpha < 1$. This situation is illustrated in Fig. 7 for alternation $\alpha = 0.8$ and Fig. 8 for alternation $\alpha = 0.5$, respectively. In each figure comparison dispersion curves (exact in this case) and the corresponding finite-N states are shown for the spin-$\frac{1}{2}$, alternating XY model. The similarities between the two models are very striking; for example, the pattern of convergence to the infinite-N limit. This gives us confidence that our numerical extrapolations are reliable.

In Fig. 9 we show the results of extrapolations on branch β_2 as a function of alternation α. Actually, the extrapolations are of the energy difference between the ground state and the lowest state of β_2. Curves for finite $N = 2$, 4, 6, 8, 10, and 12 are also shown. This figure is an extended version of Fig. 5 of Ref. 15 (Duffy and Barr). Clearly, for $\alpha \lesssim 0.5$, the convergence is very rapid, and the existence of an energy gap is not in doubt. For $\alpha \lesssim 1.0$, however, the convergence is much poorer and there is definite uncertainty in our extrapolations, affecting the actual magnitude of the energy gap near the uniform limit. However, the extrapolation procedures clearly indicate the existence of a gap for $\alpha < 1$ and *not* $\alpha = 1$. In Fig. 10 we show the results of extrapolations on the lowest states of branch β_1. Again, the energy gaps for $N = 4$, 6, 8, 10, and 12 are also shown. The convergence is rather slower in this case than in the case of branch β_2. However, as the figure shows, the extrapolation results for β_1 are very close to those for β_2, and may be regarded as identical, to within the

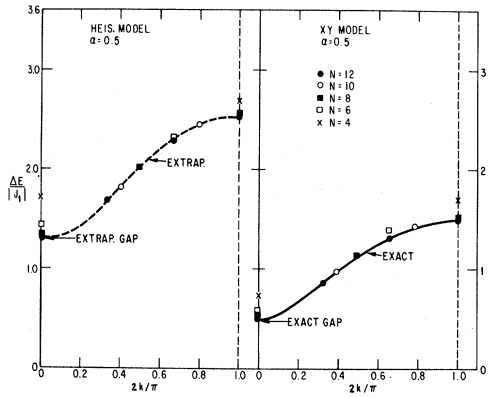

FIG. 8. A comparison figure to Fig. 7, corresponding to alternation $\alpha = 0.5$.

limits of numerical uncertainty. Hence we infer that branch β_1 becomes degenerate with branch β_2 in the limit $N \to \infty$ in the alternating case as well as the uniform case.

A further set of spectral excitations which might possibly show a gapless or acoustic character in the $N \to \infty$ limit, is the alternating equivalent of the singlet dispersion branch γ in Fig. 5. As already noted, branches γ and β become degenerate (and gapless) as $N \to \infty$ for the uniform chain. There would be the possibility that this $N = \infty$ singlet-triplet degeneracy is broken for finite alternation, with the singlet dispersion branch remaining gapless and the triplet branch showing a gap. This is

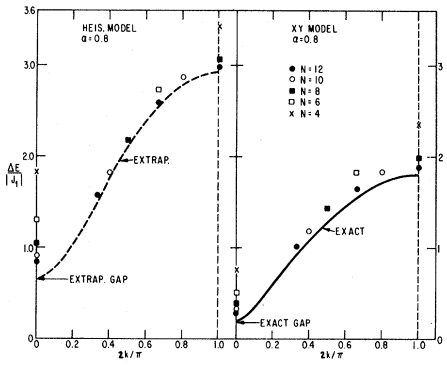

FIG. 7. A comparison of the lowest-lying triplet branches for alternation $\alpha = 0.8$, extrapolated from finite-N calculations for the Heisenberg antiferromagnet (dashed curve) and exact for the XY model (solid curve). The corresponding finite-chain states are shown in both cases.

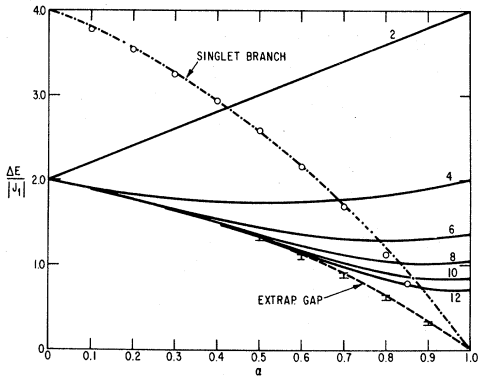

FIG. 9. Extrapolation (dashed curve) as a function of alternation α of the energy difference between the ground state and the lowest state of optic dispersion branch β_2. Curves for finite $N = 2$, 4, 6, 8, 10, and 12 are shown also. The dotted-dashed curve represents extrapolations on the minimum singlet energy gap.

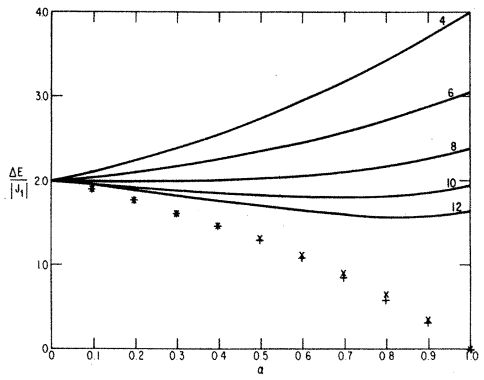

FIG. 10. Extrapolated points (denoted +) of the energy differences between ground states and lowest states of the acoustic dispersion branch β_1. Corresponding curves for finite $N=4$, 6, 8, 10, and 12 are shown also. For comparison, the optic extrapolated points (denoted $\times$) from Fig. 9 are displayed. The close agreement of extrapolations performed on two distinct classes of states is very significant, implying that both branches β_1 and β_2 become degenerate (optic) as $N\to\infty$.

an interesting possibility, since nonmagnetic singlet states would not qualitatively affect magnetic susceptibility or spin-resonance experiments, nor would they contribute any spectral weight in low-temperature inelastic neutron scattering studies. Hence, existing experimental studies of spin-Peierls and spin-exciton systems which indicate a triplet gap would not necessarily be in conflict with a gapless, singlet acoustic mode.[65] Accordingly, extrapolations were performed on the lowest state of the "alternation-reflected" portion of the γ branch (see the appropriate set of finite-N states at $k=0$ in Figs. 7 or 8). The resulting curve is shown in Fig. 9. In the dimer limit, the gap corresponding to this low-lying singlet is exactly *twice* the triplet gap, and a gap which is approximately (i.e., to within numerical extrapolation accuracy) twice the triplet gap persists apparently to $\alpha=1$, where it vanishes. This result does indeed demonstrate that the singlet-triplet degeneracy in the $N\to\infty$ uniform limit is broken in the presence of alternation, but in such a way that the singlet branch lies above the triplet branch, not below, and therefore the singlet branch does not yield a gapless mode. Although still higher-order excitation dispersion branches may be identified in the finite-N spectra, their convergence is too poor for reasonable extrapolation. However, it is considered most unlikely that they play a significant role.

The exact finite-sequence extrapolation technique has been found to be very successful for the thermodynamic properties of the magnetic chain, in cases where comparison can be made with a reliable alternative numerical calculation based on an exact analytic formalism.[38] However, it seemed to us worthwhile to check the extrapolation technique on two exactly solvable one-dimensional systems showing energy gaps, both gaps vanishing in a particular limit. These two models are the spin-$\frac{1}{2}$, alternating XY model whose Hamiltonian is

$$H_{xy}=2|J_1|\sum_{i=1}^{N/2}(S^x_{2i-1}S^x_{2i}+S^y_{2i-1}S^y_{2i}) +2|J_2|\sum_{i=1}^{N/2}(S^x_{2i-1}S^x_{2i}+S^y_{2i}S^y_{2i+1}) , \tag{2.3}$$

and the antiferromagnetic, spin-anisotropic Ising-Heisenberg model with uniform coupling:

$$H_{I-H}=2|J|\sum_{i=1}^{N}[S^z_iS^z_{i+1}+\gamma(S^x_iS^x_{i+1}+S^y_iS^y_{i+1})] . \tag{2.4}$$

In the latter case, it should be mentioned that numerical extrapolations (on rings up to 10 spins only) were made[6] *before* an analytic result appeared.[39,40] Comparison[20] shows that the early numerical extrapolations correctly predicted that the energy gap vanished in the limit $\gamma\to1$. The analytic result is that the gap, in fact, vanishes with an essential singularity, in qualitative agreement with the numerical results.

We have repeated the numerical extrapolations on larger rings and comparison with the analytic result is shown in Fig. 11, along with the curves for finite $N=2$, 4, 6, 8, 10, and 12 spins. Again, the extrapolations indicate that the gap vanishes only in the isotropic limit $\gamma=1$. In the region $\gamma\lesssim0.5$, convergence is more rapid and the extrapolations are quantitatively rather accurate. Greater quantitative uncertainty arises in the region $\gamma\gtrsim0.5$, but the extrapolations apparently consistently yield a lower bound.

The second system, the spin-$\frac{1}{2}$, alternating XY chain has an energy gap proportional to 1-α,[41,42,43] therefore vanishing linearly in the uniform limit $\alpha\to1$. In Fig. 12 we show the exact gap, the gaps for finite $N=4$, 6, 8, 10, and 12, and the numerical extrapolations. Again, the extrapolations reli-

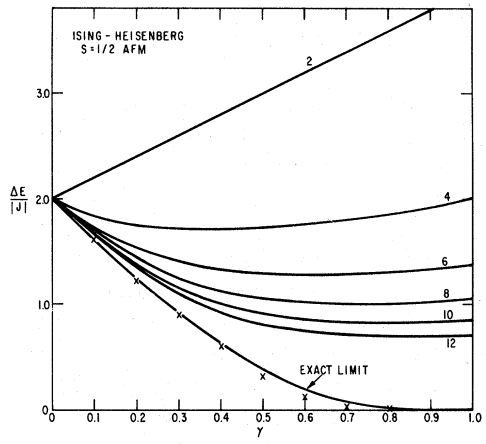

FIG. 11. Test extrapolated points (shown as crosses) on rings of $N=2$, 4, 6, 8, 10, and 12 spins for the spin-$\frac{1}{2}$, uniaxially anisotropic Ising-Heisenberg antiferromagnetic chain. The exact solution for this model is shown as the solid curve and a comparison indicates the expected accuracy of finite-chain extrapolation techniques.

ably predict that the gap vanishes as $\alpha \to 1$, and are quantitatively very accurate in the region $\alpha \lesssim 0.7$. However, we see again quantitative uncertainty near $\alpha = 1$, making it difficult to be sure of the correct functional dependence of the gap on α in this region. However, again we observe that the extrapolations consistently form a lower bound on the exact result.

Studies on these two exactly solvable test cases therefore give us considerable confidence that our extrapolation techniques (1) reliably indicate

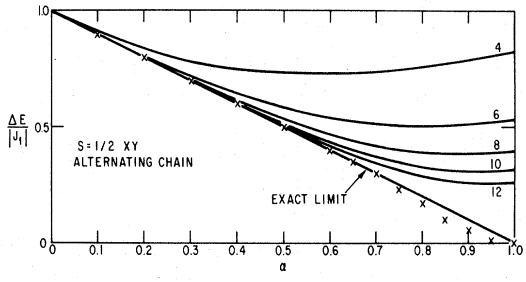

FIG. 12. Test extrapolated points (shown as crosses) on rings on $N=4$, 6, 8, 10, and 12 spins for the spin-$\frac{1}{2}$, alternating XY chain. The exact solution is shown as the solid straight line. The extrapolations generally give good agreement with the exact result, except for $\alpha \gtrsim 0.85$, where the extrapolated points lie consistently below.

whether a gap is present or not, (2) give reasonable quantitative accuracy over the whole parameter range, and (3) where some degree of quantitative uncertainty does exist, the extrapolations appear to give a lower bound on the gap. Therefore we believe our numerical studies are both exhaustive and quantitatively quite reliable, and they refute the existence of a gapless mode as predicted by Drawid and Halley.[30]

III. ANTIFERROMAGNETIC GROUND STATE

The behavior of the energy gap is not the only question of interest in the case of the alternating Heisenberg antiferromagnetic chain. A decrease in magnetic energy as dimerization takes place is a necessary condition for the existence of a spin-Peierls transition in the system. Whether a given system can possibly undergo a spin-Peierls transition therefore be determined by an examination[4] of the behavior of the $T=0$ free energy, i.e., the ground-state energy, as a function of α [or, better, the parameter $\delta=(1-\alpha)/(1+\alpha)$]. We have used techniques outlined in Appendix B to extrapolate the $E_0^{(N)}(\delta)$, i.e., the finite-N ground-state energies for various δ values, and believe our results to have good accuracy over the whole range of α (or δ). It is possible, in fact, to extrapolate the finite-N ground-state energies much more accurately than it is possible to extrapolate the energy gaps. The results are compared with those of other theories in the final section (Sec. V).

However, we should note that the alternating ground-state energy is interesting in its own right, and not merely in connection with spin-Peierls theory. In Fig. 13, the extrapolated ground-state energy E_0 is shown as a function of the two exchange constants J_1 and J_2, and consideration is given also to the case where J_1 and/or J_2 are ferromagnetic. The upper curve through the extrapolated points (circles) refers to $J_1 < 0$, i.e., antiferromagnetic, and the horizontal scale gives J_2. For example, the point at $J_2 = 0$ gives the (exactly known) energy of isolated, antiferromagnetic dimers. The extrapolated points to the right of the dimer limit correspond to ferromagnetically coupled antiferromagnetic pairs (dimers). The points behave smoothly as a function of J_2 (>0), and the point at $J_2 = 1 = -J_1$ shows, to numerical accuracy, no evidence of being a special point. However, to the left of the dimer limit where $J_2 < 0$ (antiferromagnetic), the point at $J_1 = J_2 = -1$ does show

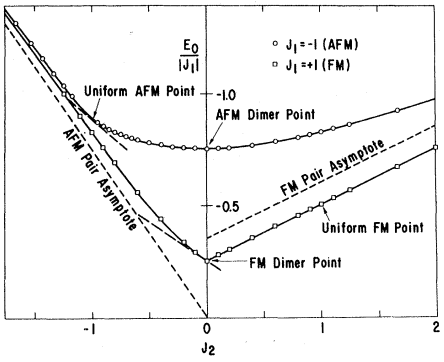

FIG. 13. Plot of the ground-state energy of alternating Heisenberg chains, both antiferromagnetic and ferromagnetic, as a function of the two exchange constants, J_1 and J_2. The significance of the asymptotes and special points is discussed in the text.

features which indicate that it is a special point (singular point). This corresponds, of course, to the uniform limit of the alternating antiferromagnet. At this point, no discontinuity is apparent in the curve or in its first derivative ($\partial E_0/\partial J_2$), but its second derivative is estimated to be divergent. The situation is analogous to the case of the antiferromagnetic ground-state energy of the Ising-Heisenberg linear chain, where again the point at $\gamma=1$ is a special point.[6,44,45] However, the singularity in this case is much weaker (an essential singularity) which may be hard to detect by numerical techniques. Returning to Fig. 13, it should be noted that if the tangent ($\partial E_0/\partial J_2$) exists at the uniform limit, it can be shown to be equal to one half the value of E_0 itself at the uniform limit. Since E_0 is known exactly,[46] it follows that the slope is also known exactly.

The lower curve through the extrapolated points (squares), refers to $J_1>0$, i.e., ferromagnetic, again plotted versus J_2 on the horizontal scale. Note that in the limits $J_2\to\pm\infty$, the upper and lower curves converge, representing isolated antiferromagnetic and ferromagnetic dimers, respectively. The point at $J_2=0$ also corresponds to isolated ferromagnetic dimers. The tangent to the left of this dimer point has a slope directly related to the ground-state energy of a spin-one, antiferromagnetic (uniform) chain.[47,48] The points to the right of the $J_2=0$ dimer limit correspond to a completely ferromagnetically coupled alternating chain. The uniform limit ($J_2=1$) shows no evidence of being a special point.

IV. CLASSICAL AND OTHER EXACTLY SOLVABLE ALTERNATING CHAINS

Our numerical studies on Heisenberg antiferromagnetic alternating chains and exact results for XY alternating chains show that an excitation energy gap exists for all nonzero alternation. However, we have remarked above that a classical calculation for the Heisenberg case[27] gives no gap. To investigate further this interesting point, we have performed a systematic study of a variety of classical systems, including, in addition to the Heisenberg alternating antiferromagnet (HAA), the XY alternating magnet (XY-A), the Heisenberg alternating ferromagnet (HAF), and an intermediate model between Heisenberg (antiferromagnetic and ferromagnetic) and XY (XY-HAA and XY-HAF). Two approaches have been used: a straightforward spin-wave approach based on classical dynamics and also a quantum-operator approach taking the limit $S\to\infty$. Both techniques yield identical results. Comparison can be made not only with the numerical extrapolations for the alternating Heisenberg, spin-$\frac{1}{2}$ antiferromagnet, but also with exact results for the spin-$\frac{1}{2}$, alternating ferromagnet model (AFM) and the spin-$\frac{1}{2}$, alternating XY model.[41–43,49]

The most striking result of these calculations is that, whereas the spin-$\frac{1}{2}$, alternating Heisenberg antiferromagnet (numerically) and the XY alternating magnet (exactly) show an excitation energy gap, the corresponding classical (spin-∞) systems do not. This represents a further demonstration that low-dimensional systems show very different behavior at low temperatures in the quantum and classical limits. It also dramatically points out the inapplicability of spin-wave theory. Furthermore, the fact that a similar situation occurs in a model exactly solvable in both limits (alternating XY), strongly supports our numerical conclusions about the Heisenberg alternating antiferromagnet.

However, an additional striking feature of these calculations, which has not, to our knowledge, been pointed out before, is the variety of excitation behavior which results. Alternating spectra are obtained which show the conventional "expected" behavior, i.e., both an acoustic and an optic branch, one model shows only acoustic excitations, and others show only optic excitations. Each model is discussed in some detail in what follows. A preliminary catalogue of the salient features of the models is presented in Table I, to help the reader avoid confusion. To predict the presence or

TABLE I. Comparative summary of the variety of excitation behavior of classical and quantum linear-spin models in relation to properties of the ground state.

Spin value	Model	LRO in ground state	Acoustic branch	Optic branch
$S=\infty$	HAA	yes	yes	no
$S=\frac{1}{2}$	HAA	no	no	yes
All S	HAF	yes	yes	yes
$S=\infty$	XY-A	yes	yes	yes
$S=\frac{1}{2}$	XY-A	no	no	yes
$S=\infty$	XY-A	yes	yes	yes
$S=\infty$	XY-HAF	yes	yes	yes

absence of an acoustic branch, the degeneracy of the ground state should be known. This represents an extension of Goldstone-mode discussions in the introduction for systems of isotropic spin symmetry to more general systems. Systems with a degenerate ground state have, in consequence, long-range order (LRO) in the ground state or, equivalently, have broken symmetry in the ground state. Spin-wave theory correctly gives the number of gapless modes, depending on the symmetry. In quantum systems with no broken symmetry, spin-wave theory is inapplicable and gapless, acoustic modes are absent. The lack of an optic branch in one of the models ($S=\infty$, HAA) has also to be explained. An interpretation in the context of a classical dimer (spin-pair) model is given subsequently.

Specifically, the classical expression for the Heisenberg AFM is

$$\frac{\Delta E}{J_1 S}=4\sqrt{\alpha}\,|\sin k|\ . \tag{4.1}$$

In the uniform limit $\alpha=1$, this expression reduces to the well-known spin-wave result[50]:

$$\frac{\Delta E}{J}=2\,|\sin k|\,,\quad (S=\tfrac{1}{2})\ . \tag{4.2}$$

It is noteworthy that the classical system shows only gapless branches, whereas we find only branches with a gap in the corresponding spin-$\frac{1}{2}$ system (see Figs. 14 and 15, respectively[51]) for general α. However, the absence of an optical branch can be understood by considering a dimer of two strongly coupled antiferromagnetic spins, with only a weak interaction with other pairs. Applying classical mechanics, one may see that (in contrast to the ferromagnetic and XY cases below) the dimer does not perform small-amplitude oscillations by itself about an equilibrium position. Interactions with other dimers are therefore essential. It has also been noted,[11] on the other hand, that the behavior of the quantum-AFM dimer completely violates the fundamental premise of spin-wave theory that the ground state is the doubly degenerate Néel state, even for $\alpha=0$. The antiferromagnetic Heisenberg dimer is, of course, the well-known singlet-triplet system, where the ground state is a singlet and the excited state is a (degenerate) $S=1$ triplet. This gives some physical insight into the contrasting behavior of the quantum and classical systems.

The comparable result for the alternating Heisenberg ferromagnetic chain is

$$\frac{\Delta E}{J_1 S}=2(1+\alpha)[1\pm(\cos^2 k+\delta^2\sin^2 k)^{1/2}]\ , \tag{4.3a}$$

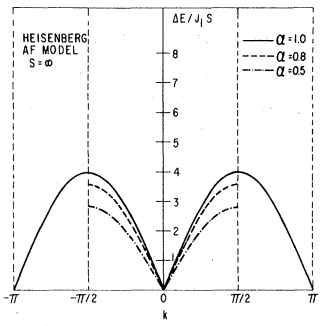

FIG. 14. Dispersion curves for the classical (spin-∞), Heisenberg antiferromagnetic chain for $\alpha=1$ (uniform), $\alpha=0.8$, and $\alpha=0.5$. Note that only gapless (acoustic) modes are present.

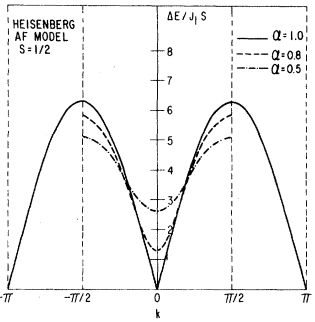

FIG. 15. Comparison figure to Fig. 14. Dispersion curves for the spin-$\frac{1}{2}$ Heisenberg antiferromagnetic chain for $\alpha=1$ (uniform), $\alpha=0.8$, and $\alpha=0.5$. Note that by contrast with the classical counterpart, for $\alpha<1$ only excitations with a gap (optic) occur.

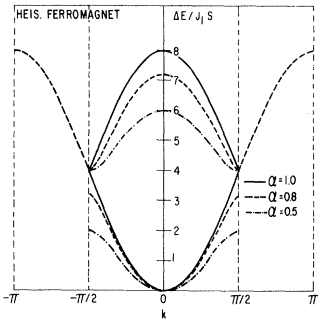

FIG. 16. Dispersion curves for the Heisenberg ferromagnetic chain, valid for all spin values, for $\alpha=1$ (uniform), $\alpha=0.8$, and $\alpha=0.5$. Note that for the ferromagnetic cases (for $\alpha<1$), both gapless (acoustic) and optic modes are present.

where

$$\delta=\frac{1-\alpha}{1+\alpha}$$

and

$$J=\tfrac{1}{2}(J_1+J_2)=\tfrac{1}{2}(1+\alpha)J_1 .$$

Expression (4.3a) may also be written

$$\frac{\Delta E}{J_1 S}=2(1+\alpha)\pm 2(1+\alpha^2+2\alpha\cos 2k)^{1/2} . \tag{4.3b}$$

The expression holds for all S, *including the case* $S=\frac{1}{2}$ (which can easily be calculated exactly by quantum mechanics), and this result establishes some justification for the spin-wave approximation for Heisenberg ferromagnets.

In the uniform limit, expression (4.3) becomes

$$\frac{\Delta E}{JS}=4(1\pm\cos k)=\begin{cases}8\sin^2 k/2 & \text{or}\\ 8\cos^2 k/2 .\end{cases} \tag{4.4}$$

For general α, two types of excitations are present, an acoustic (gapless) mode and an optic mode with an excitation gap. This result is illustrated in Fig. 16 for several values of α.

The classical result for the alternating XY linear chain is

$$\frac{\Delta E}{J_1 S}=2(1+\alpha)[1\pm(\cos^2 k+\delta^2\sin^2 k)^{1/2}]^{1/2} , \tag{4.5}$$

where again,

$$\delta=\frac{1-\alpha}{1+\alpha}$$

and is shown in Fig. 17 for various α values.

In the uniform limit, (4.5) reduces to

$$\frac{\Delta E}{JS}=4(1-\cos k)^{1/2}=4(2)^{1/2}\,|\sin k/2| . \tag{4.6}$$

Again, for general α, we see two types of excitations, acoustic (gapless) and optic. In this respect, the classical alternating XY chain differs strikingly from the classical alternating Heisenberg antiferromagnetic chain, which has no optic branch, as discussed above. Accordingly, we shall subsequently investigate how the classical optic branch makes its appearance on proceeding from the Heisenberg to the XY limits. The other feature of considerable interest concerning the XY alternating chain is the difference between the $S=\infty$ and $S=\frac{1}{2}$ limits. The exact $S=\frac{1}{2}$ solution is shown in Fig. 18, and is given by the expression

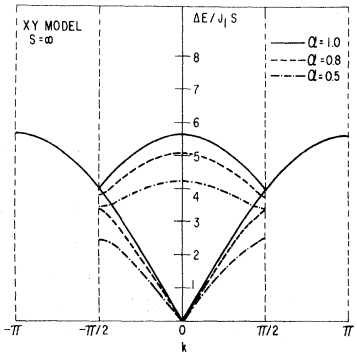

FIG. 17. Dispersion curves for the classical (spin-∞) XY chain for $\alpha=1$ (uniform), $\alpha=0.8$, and $\alpha=0.5$. For $\alpha<1$, both acoustic and optic modes are present.

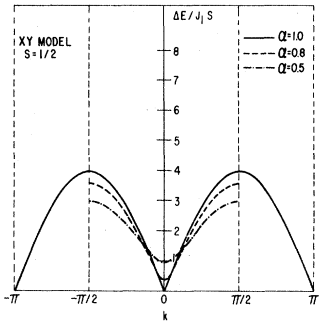

FIG. 18. Comparison figure to Fig. 17. Dispersion curves for the spin-$\frac{1}{2}$ XY chain for $\alpha=1$ (uniform), $\alpha=0.8$, and $\alpha=0.5$. Note that for the spin-$\frac{1}{2}$ case, only optic modes are present for $\alpha<1$ and the situation qualitatively resembles that of the spin-$\frac{1}{2}$, alternating Heisenberg antiferromagnet.

$$\frac{\Delta E}{J_1 S}=2(1+\alpha)(\sin^2 k+\delta^2\cos^2 k)^{1/2}\,, \tag{4.7}$$

where

$$\delta=\frac{1-\alpha}{1+\alpha}\,.$$

No acoustic (gapless) branch appears in this expression for $\alpha<1$, and the excitation energy gap is given by

$$\frac{\Delta E}{J_1}=1-\alpha \tag{4.8}$$

(see Fig. 12). The excitation spectrum qualitatively resembles that of the spin-$\frac{1}{2}$, Heisenberg linear alternating chain (see Fig. 15). Finally, in Figs. 19 and 20, we investigate the classical dispersion spectra as the spin anisotropy varies. For convenience, we consider the limit $\alpha=1$, but show a reduced zone appropriate to an alternating system. Starting from the XY limit and proceeding to the Heisenberg antiferromagnet we observe the following features of Fig. 19: (1) The acoustic branch of the XY model is modified very little as the Heisenberg limit is approached, but (2) the optic branch is strongly affected, dipping down, and eventually becoming degenerate with the acoustic branch in the Heisenberg limit. Similar behavior is observed for other alternation (α) values. The dispersion relation for general alternation is given by

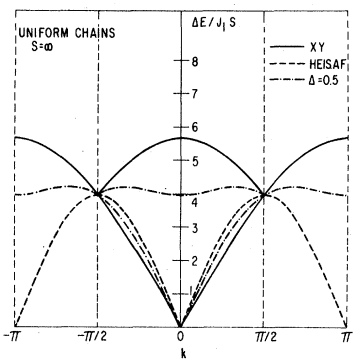

FIG. 19. Variation of classical dispersion spectra (considering only the uniform limit for convenience) with anisotropy between the XY limit and the antiferromagnetic Heisenberg limit. Note that the acoustic branch is little affected, but the optic branch is strongly affected, dipping down and becoming degenerate with the acoustic branch in the Heisenberg limit.

$$\frac{\Delta E}{J_1 S}=2(1+\alpha)[1-\phi(\cos^2 k+\delta^2\sin^2 k)\pm(1-\phi)(\cos^2 k+\delta^2\sin^2 k)^{1/2}]^{1/2}\,, \tag{4.9}$$

where

$$\delta=\frac{1-\alpha}{1+\alpha}$$

and

$$0\le\phi\le 1\,,$$

where

$$\phi=J_z/J_x,\quad J_x=J_y\,.$$

$\phi=0$ corresponds to the XY model and $\phi=+1$ corresponds to the antiferromagnetic Heisenberg model. In the uniform limit, expression (4.9) becomes

$$\frac{\Delta E}{JS}=4[(1-\cos k)(1+\phi\cos k)]^{1/2}\,. \tag{4.10}$$

For the less dramatic case of the classical XY-Heisenberg ferromagnet, the dispersion relation is simply given by (4.10) with the sign of ϕ reversed. The variation between the XY limit ($\phi=0$) and the Heisenberg ferromagnet ($\phi=-1$) is shown in Fig. 20.

In summary, we have examined a number of exactly solvable alternating linear systems and found

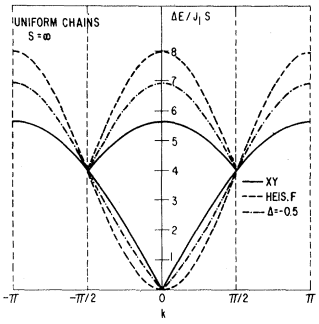

FIG. 20. Companion figure to Fig. 19. Here variation of classical dispersion curves is shown between the XY limit and the ferromagnetic Heisenberg limit.

much diversity in their excitation spectra. Only the ferromagnetic Heisenberg system displays the "expected" behavior for an alternating system, namely both an acoustic and an optic mode, for all values of spin S. The XY model shows both an acoustic and optic mode in the classical limit, $S=\infty$, but shows only an optic mode for $S=\frac{1}{2}$. These are both exact results, and on the surface indicate a breakdown of spin universality for the critical singularities at $T=0$ and also a breakdown of spin-wave theory for the quantum limit. The alternating Heisenberg model shows only acoustic modes in the $S=\infty$ limit and, according to our numerical studies, only optic modes in the $S=\frac{1}{2}$ limit, again in apparent violation of spin universality. We were therefore stimulated to investigate the case of intermediate spin S. For the alternating antiferromagnetic chain with $S=1$, our finite-ring extrapolations show gap values very close in magnitude (as a function of α) to the corresponding gap-value extrapolations for $S=\frac{1}{2}$. It is therefore tempting to infer that the gap remains essentially independent of S. However, the normalization of energies required when proceeding to the $S\rightarrow\infty$ limit implies a gap vanishing as $\Delta E/S$. Since $\Delta E\times S$ is then effectively constant, independent of S, spin universality, from this perspective, is preserved.

V. DISCUSSION

Our primary interest in this section is to discuss the probable accuracy of our numerical results for E_0, the ground-state energy per spin of the Heisenberg alternating antiferromagnetic chain as a function of alternation, and ΔE, the corresponding excitation energy gap between the singlet ground state and the triplet lowest-lying excited states. The discussion will focus on both the overall quantitative accuracy of our results over the entire range of alternation and the specific nature of the behavior of both E_0 and ΔE near the uniform limit, which is important for spin-Peierls theory.[4] Our discussion will include a comparative assessment of our results in relation to those of other approximate theories.

Let us first consider the ground-state energy. To investigate the behavior near the uniform limit it is appropriate to consider the deviation of the energy for nonzero alternation, $E_0(\delta,J)$, from its value at the uniform limit, $E_0(0,J)$. Hence we define the quantity

$$\epsilon(\delta)=E_0(0,J)/J-E_0(\delta,J)/J\ .$$

We give predictions for the asymptotic behavior of $\epsilon(\delta)$ as $\delta\rightarrow 0$ for the various theories and models. For the XY model (exact) $\epsilon(\delta)\sim\delta^2\ln\delta$ (Ref. 49), and for the Hartree-Fock approximation[11] to the Heisenberg case, $\epsilon(\delta)\sim\delta^2\ln^2\delta$. The Luttinger-model approach of Cross and Fisher gives rise to a different functional dependence on δ, i.e., a pure power law, $\epsilon(\delta)\sim\delta^{4/3}$. Let us first make a rough assessment of which functional form is in best accordance with our numerical results for the alternating Heisenberg chain. In Fig. 21 our extrapolated data points near the uniform limit are plotted in terms of $f(\delta)$, where $f(\delta)$ takes on the three forms above. Agreement with a particular functional form will result in a straight-line plot.

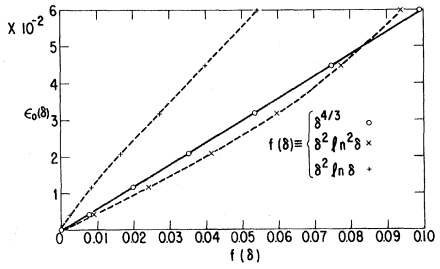

FIG. 21. Extrapolated numerical data points for the ground-state energy of the alternating Heisenberg antiferromagnetic chain plotted in terms of three functional forms: the Cross-Fisher theory (points denoted ○), the Bulaevskii Hartree-Fock theory (points denoted ×), and the XY model (points denoted +). The plot in terms of Cross-Fisher theory gives the best straight line.

Clearly, agreement (at least within numerical accuracy) of the Heisenberg data with the expression for the XY model is quite poor, as we should expect. The Bulaevskii Hartree-Fock prediction is in much better agreement for small δ, but significant deviations appear for $\delta \gtrsim 0.07$. The fit to the Cross-Fisher prediction is strikingly good. The special features of the Cross-Fisher calculation for alternating chains, which is substantially based on earlier work by Luther and Peschel[25] on uniform chains, have already been discussed at some length by these authors. Here we shall just give the relevant highlights. The Luther-Peschel continuum (Luttinger) approximation applied directly gives exact $T=0$ critical exponents for the XY model but results which are approximate to the extent of a few percent for the antiferromagnetic Heisenberg model. However, Cross and Fisher have made use of a valuable feature of the model, namely that scaling relations are preserved between exponents. Hence if an exact exponent is fed in, additional exponents are obtainable which should also be exact. This is the basis on which the Cross-Fisher 4/3 exponent was obtained.

Our numerical results of Fig. 21 lend support to power-law behavior. Another feature of interest, observed first by Cross and Fisher, is that the 4/3 power law appears to be valid not only in the asymptotic region $\delta \to 0$, but over the whole range of dimerization $0 < \delta \leq 1$. Cross and Fisher used older data of Duffy and Barr. Using our more recent, extensive data on longer chains, we have apparently verified their observation. In Fig. 22 a log-log plot of 14 data points yields an apparently linear behavior with a "best exponent" of $1.36^{+0.1}_{-0.2}$. There is no obvious reason why power-law behavior close to 4/3 should extend right to the dimer limit. This result is at present a curiosity (a 4/3 power law over the whole alternation range is not consistent with the Harris expansion[17]). In fact, we shall see, in view of subsequent theoretical considerations, that pure power law is *not* the expected behavior near the uniform limit.

The $T=0$ quantum-renormalization-group (QRG) approach also predicts values for exponents. In view of the rather high current level of interest in this particular approach, we shall review it separately, later in this section.

We now concern ourselves with the overall quantitative accuracy of the various calculations of $E_0(\delta,J)$. A specific inherent feature, and unfortunate limitation, of the Cross-Fisher calculation is that the nature of the critical singularities is obtainable, but unique values for the corresponding amplitudes and therefore actual values for $E(\delta,J)$ (or ΔE) cannot reasonably be obtained. Hence it is difficult to make comparison with our extrapolations. In Fig. 23 our extrapolations (solid curve) are compared with the Bulaevskii Hartree-Fock ap-

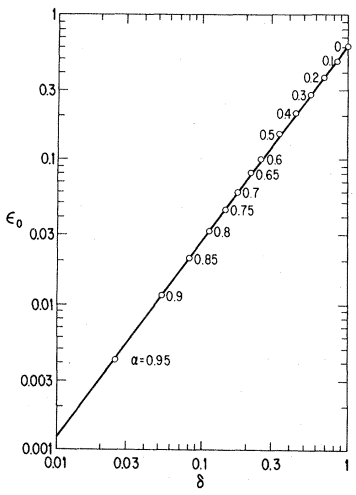

FIG. 22. A log-log plot of 14 numerically extrapolated Heisenberg data points for the alternating ground-state energy. A very good straight-line plot with a slope of ~ 1.36 is obtained over the entire alternation range.

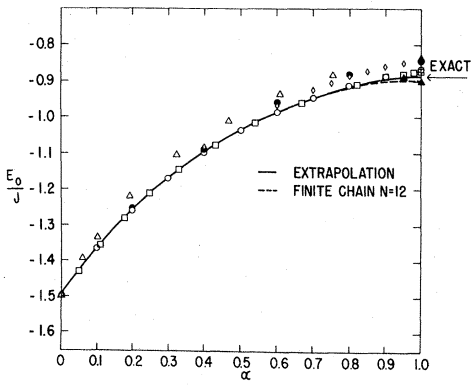

FIG. 23. Comparison of ground-state energy per spin as a function of alternation for various models. Open triangles denote points corresponding to the Bulaevskii Hartree-Fock theory. Open squares come from the theory of Klein and Garcia-Bach and open circles from a theory of Harris. Solid circles denote an $N_L=2$, $N_S=9$ RG calculation of Fields and open diamonds an $N_L=4$, $N_S=4$ RG calculation. Points from a quasiboson theory of Montgomery are shown as solid triangles where they deviate from the extrapolated results (solid curve) near the uniform limit. The finite-chain $N=12$ curve is shown dashed where it deviates from the $N \to \infty$ extrapolated result, again near the uniform limit.

proximation and an appreciable quantitative discrepancy is observed for all alternation. Note that the exact result of Hulthén,[46] for the uniform limit only, is indicated by the arrow. It has already been shown[6] that extrapolations in terms of $1/N^2$ give very high accuracy in this limit. Since the dimer limit is given trivially and exactly for all finite N, and our extrapolations give excellent agreement in the uniform limit, there is good reason to believe that for intermediate alternation our extrapolated results should be quite accurate. A Kekulé state calculation of Klein and García-Bach[52] gives much better agreement with our extrapolations than the Bulaevskii calculations. A discrepancy between our results and this valence-bond-type calculation appears only very close to the uniform limit, where there is a 1.5% discrepancy. It may be remarked that this Kekulé state approach has also been applied to the spin-1 alternating chain,[52] where it is less successful, showing a 12% discrepancy with accurate extrapolations for the ground-state energy in the uniform limit.[47,48] We consider this to result from the fact that the valence-bond approach is inherently more suited to an $S=\frac{1}{2}$ rather than an $S=1$ alternating problem. Finally, in Fig. 23, we observe that a third-order perturbation-theory calculation of Harris,[17] evaluated over the entire range of alternation, also gives very good results. This is interesting, and perhaps surprising, since the calculation is a perturbation calculation about the dimer limit.

We come now to the case of the energy gap ΔE. Again, specific predictions are available for the way in which ΔE vanishes in the vicinity of the uniform limit. For the XY model, $\Delta E \sim \delta$ (exact), whereas for the Bulaevskii approximation to the Heisenberg model $\Delta E \sim \delta \ln \delta$. A quasiboson calculation of Montgomery gives $\delta^{1/2}$. The Cross-Fisher theory predicts $\delta^{2/3}$. Unfortunately, our extrapolations are not sufficiently accurate close to the uniform limit to yield an exponent. The only conclusion we can reasonably draw is that the curvature indicates an exponent less than unity. Very recent work has given us an insight into why convergence of numerical approaches is slow, and will be discussed subsequently.

Regarding quantitative accuracy, a comparison of various approximate theories for the alternating excitation energy gap is shown in Fig. 24. This time we have included a Cross-Fisher-based calculation of ΔE. The normalization is arbitrary and the form $\delta^{2/3}$ is assumed to hold over the entire range of δ, by analogy with observations on E_0.

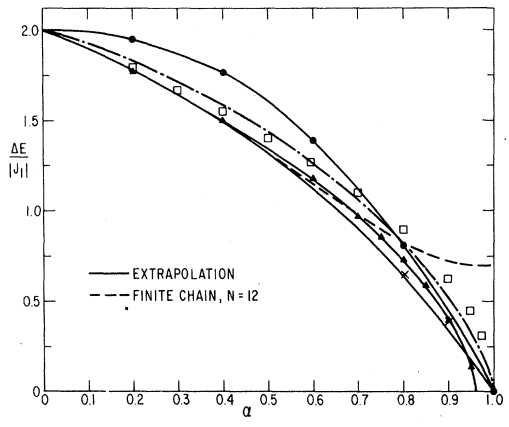

FIG. 24. Comparison of various approximate theories for the alternating Heisenberg anitferromagnetic excitation energy gap. The direct extrapolations are shown as a solid curve without symbols. The $N=12$ curve is shown dashed where it departs from the limiting curve. The solid curve through the solid circles is the $N_L=2$, $N_S=9$ RG calculation, whereas the solid curve through solid triangles is the $N_L=4$, $N_S=4$ RG calculation. The open squares represent a quasiboson calculation of Montgomery. The dashed-dotted line gives a Cross-Fisher-type, $\frac{2}{3}$ power-law expression with arbitrary normalization. The extrapolations are in remarkably good agreement with the Bulaevskii theory (points shown by curves), deviating only for $\alpha \geq 0.8$ near the uniform limit.

Our extrapolated results are shown with the $N=12$ curve for comparison. For $\alpha[=(1-\delta)/(1+\delta)] \lesssim 0.5$, convergence has effectively occurred. Theories which agree with our results in this region, where they may be regarded as quantitatively reliable, include the perturbation calculation of Harris[17] and the Hartree-Fock approach of Bulaevskii.[11] In fact, the Bulaevskii results are remarkably close to our direct extrapolations over the whole alternation range. This result gives *ex post facto* justification for use of a theory incorporating a Hartree-Fock approximation to interpret recent experimental results as indicating the first good example of a spin-Peierls system.[3,4] The quasiboson calculation shows deviations from the extrapolations at all alternation values, including the vicinity of the dimer limit, and must therefore be regarded as less reliable than other theories. For the energy gap, the Harris perturbation results are only reliable for $\alpha \lesssim 0.5$. Attempts to continue to the uniform limit, whether by direct evaluation or more sophisticated Padé approximant techniques, tend to result in persistence of the gap even

at the uniform limit, a result which is well known to be incorrect.

An abbreviated discussion of some of these results and comparisons has already appeared in the context of $T=0$ quantum-renormalization-group (QRG) theory.[32] The $T=0$ QRG is a variant of the real-space renormalization-group approach pioneered by Niemeijer and van Leeuwen[53] which focuses on the ground state and dominant set of lowest-lying excited states.[54,55] It is therefore particularly suitable for treating ideal one-dimensional (1D) systems whose critical singularities occur at $T=0$, and also systems characterized by a gap which vanishes at a particular value of a variable parameter in the Hamiltonian. Hence, it has been used to investigate $E_0(\delta)$ and $\Delta E(\delta)$ for $S=\frac{1}{2}$ XY and Heisenberg antiferromagnetic alternating chains,[31-33] $S=1$ Heisenberg antiferromagnetic alternating chains,[33] and Ising-Heisenberg antiferromagnetic, uniform chains.[33] Since the $S=\frac{1}{2}$ alternating XY and the $S=\frac{1}{2}$ Ising-Heisenberg uniform models are exactly solvable,[37,39-43] they have been used to test the $T=0$ QRG method, as indeed, they have been used to check the accuracy of the direct finite-chain extrapolations (see Sec. II). Since reasonable qualitative success results in the case of the solvable test models, we shall summarize here the corresponding $T=0$ QRG results for the alternating Heisenberg antiferromagnet, and then make comparison with the extrapolations and other methods, particularly the Cross-Fisher calculation.

The essence of the method is that the lattice is divided into blocks of spins whose eigenvalues and eigenvectors may be calculated exactly. A given number of low-lying eigenstates are retained to write the interblock interaction, and the scheme is repeated until convergence results to a "fixed point." Denoting the number of spins in the block by N_s, and the number of levels retained at each iteration step by N_L, a comprehensive investigation has been undertaken, systematically increasing both N_s and N_L for both $S=\frac{1}{2}$ and $S=1$. The eigenstructure yielded by the $T=0$ QRG has been carefully compared with the eigenstructure in the thermodynamic limit as deduced from calculations on large finite-ring systems. A full report is in preparation.[33]

The QRG calculations fall naturally into two classes: N_s odd, with $N_L=2$, and N_s even, with $N_L=4$ (6 or 8). The N_s-odd calculations have the advantage that an important symmetry of the alternating Hamiltonian, namely invariance with respect to relabeling of J_1 and J_2 (or replacing α by α^{-1}), is automatically preserved. The problem is that retaining only two levels means that important information fails to be utilized at each step. It turns out that the lowest-order N_s-odd calculation, namely $n_s=3$, $N_L=2$, is equivalent to an early calculation of de Braak *et al.*[56]

When N_s is even, retention of more levels, namely $N_L=4$, allows the fundamental singlet-triplet level structure of the alternating Hisenberg chain to be preserved, and hence, offers the likelihood of greater quantitative accuracy. Unfortunately, the situation when N_s is even inherently violates J_1, J_2 symmetry.

RG calculations have been performed for $N_s=3$, 5, 7, and 9 and $N_L=2$ for the alternating Heisenberg system. Fixed points are found at specific values of α. The uniform limit, $\alpha^*=1$, is always found to be an unstable fixed point of the system. Initial values $\alpha^{(0)}<1$ flow (with increasing number of iterations n) into the stable fixed point $\alpha(n \rightarrow \infty)=\alpha^*=0$ corresponding to independent dimers. This result leads immediately to the conclusion that the alternating spectrum has a gap which vanishes only in the uniform limit. This conclusion is supported by similar fixed-point behavior in the case of the exactly solvable alternating XY model[31] and the Ising-Heisenberg uniform spin-anisotropic model,[33] which also show gaps which vanish only in the uniform (isotropic) limit. In fact, the antiferromagnetic fixed point corresponds to $\alpha^*=\gamma^*=1$, and is therefore *doubly* unstable (with respect to both alternation and anisotropy). We conclude that these RG results are in agreement with the direct finite-chain extrapolations, and thus support the existence of a gap for $\alpha<1$.

The $T=0$ QRG calculations also give exponents for $\epsilon(\delta)$ and $\Delta E(\delta)$ as $\delta \rightarrow 0$. The $N_s=3$, $N_L=2$ calculation, which can be done analytically[56] yields the result $\epsilon(\delta) \sim \delta^{1.37744}$, in very good agreement with the extrapolation exponent value of 1.36 and the Cross-Fisher value of 1.33. For larger values of N_s an analytic RG calculation is no longer feasible. The calculation must be done on a computer, and the exponent determined numerically.[57] An analysis of numerical results for $N_s=7$, $N_L=2$ near the uniform limit suggests a higher value for the exponent of ~ 1.5, which agrees with results obtained by an RG-based finite-size scaling approach.

For the sequence where N_s is even, for example, $N_s=4$, 6, and 8, it should be mentioned that the

fixed-point structure is not as satisfactory as for the N_s-odd sequence, since the unstable fixed point is no longer precisely at unity. The deviation, however, is never more than a few percent and is directly attributable to the breakdown of J_1, J_2 symmetry noted above. An investigation in the vicinity of the unstable fixed points ($\alpha^*=0.962$, for $N_s=N_L=4$) yields a value for the ϵ_0 exponent which is the same, to numerical accuracy, for all N_s, and is 1.6. This value is significantly larger than the finite-chain value and the Cross-Fisher value. Reasons for this discrepancy will be discussed later in this section.

We return to Fig. 23 to consider the quantitative accuracy of the RG approach. Clearly, global accuracy for the RG calculations under discussion is not high, a feature which tends to characterize many types of RG treatments. For the ground-state energy, the $N_L=2$, $N_s=9$ RG calculation is almost equivalent in accuracy to the $N_L=4$, $N_s=4$ RG calculation, and both are comparable with the Bulaevskii results, and hence are poorer than other calculations. Specifically, there is a quantitative discrepancy amount to 12% in the uniform limit. However, in Fig. 25 we show the $N_L=2$ RG sequence, $N_s=3$, 5, 7, and 9, extrapolated to $N_s=\infty$. The accuracy is now much improved. In Fig. 24 the RG predictions for the energy gap, ΔE, are shown. The points for the $N_L=2$, $N_s=9$ RG are seriously in error near the dimer limit (this RG fails to reproduce even the linear term of a perturbation series about the dimer limit). The $N_L=4$, $N_s=4$ RG gives much improved results, especially near the dimer limit, since the basic singlet-triplet structure is preserved at each step of the iteration

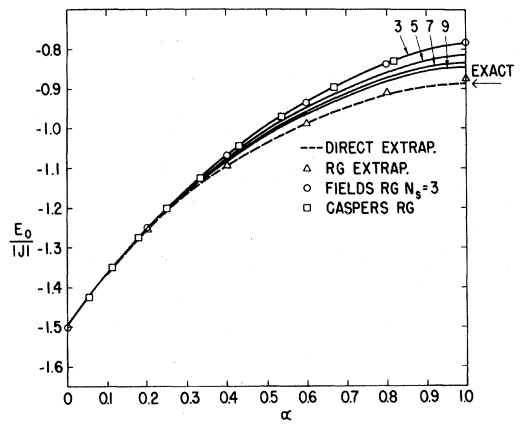

FIG. 25. Extrapolations of two-level RG groups for $N_s=3$, 5, 7, and 9 in comparison with direct numerical extrapolations on finite chains.

scheme, i.e., the symmetry is correct. Unfortunately, the $N_L=4$, $N_s=4$ gap vanishes prematurely because of breakdown of J_1, J_2 symmetry. Regarding the energy-gap exponent, the N_s-even RG calculations give an exponent of 0.62, whereas the $N_L=2$, $N_s=7$ (Ref. 31) calculations give ~ 0.76. Again there is agreement with an RG-based finite-size scaling approach, which gives an exponent of ~ 0.75.

In summary of the above, the direct finite-chain extrapolation technique appears to yield the most informative and reliable results overall. Its most serious competitors are the Cross-Fisher calculation and the $T=0$ RG calculations. Since the Cross-Fisher method cannot give amplitudes, serious quantitative comparison of spectral properties in general is precluded. The $T=0$ quantum-RG method, handled with caution, can be made to yield reasonable results for both quantitative behavior and exponents. Hence, an attempt was made to combine quantum-RG concepts with finite-size scaling techniques.[58,59] The attempt appeared rather successful, giving exact results when tested on the alternating XY model. For the Heisenberg alternating model the exponents for ϵ and ΔE have already been noted to be 1.5 and 0.75, respectively, significantly different from the Cross-Fisher values of 4/3 and 2/3, respectively. The discrepancy is a matter of some note, since the Cross-Fisher results are claimed to be exact. Very recent work has given us insight into a reason for the discrepancy between finite-size scaling exponents and Cross-Fisher exponents and also a reason for the slow convergence of the direct finite-chain extrapolations close to the uniform limit. The alternating Heisenberg antiferromagnetic chain can be mapped into a rather general kind of two-dimensional (2D) staggered eight-vertex model, equivalent to a Potts model with number of components, $q=4$.[60] Now $q=4$ is the "marginal component dimensionality" at which the 2D Potts-model transition changes from first to second order, and logarithmic factors are observed as critical corrections to scaling. The inference is that logarithmic factors may well affect (a) the convergence of the direct finite-Heisenberg-chain extrapolations close to the uniform limit, and (b) the apparent exponents derived from finite-size scaling techniques. A further, more dramatic inference, is that there are logarithmic corrections to the power-law exponents of Cross and Fisher. A future project, therefore, is to reanalyze the finite-chain results in such a manner as to take account

of logarithmic corrections, and improve accuracy near the uniform limit. Clearly the very interesting matter of the spin-Peierls critical exponents is not yet satisfactorily resolved, either numerically or in terms of an analytic (Cross-Fisher-type) calculation.[61] Note that the effect just described is an indication that the uniform limit of the Heisenberg chain behaves as a singular point, as has already been observed in another context and described in Sec. III. The uniform limit of the alternating XY chain is not a special point and the finite-size scaling technique gives exact results (as does the Cross-Fisher approach).

This work has been concerned with the low-lying spectral excitations of quantum, antiferromagnetic, alternating Heisenberg chains. From the point of view of practical application, however, static thermodynamic properties are important, and the dynamics of such systems in zero and nonzero magnetic fields would be interesting.[10] We have investigated the zero-field susceptibility, specific heat, and the zero-temperature magnetization isotherms. Our susceptibility results are already published as part of a comparative study of the Bulaevskii Hartree-Fock approximation and direct finite-chain extrapolations.[21] Extrapolations for specific heat and magnetization isotherms will be published in due course,[23] and, in the meantime, are available on request. In general, our results are in agreement with earlier studies of Duffy and Barr on rings up to 10 spins for the alternation range $0 \leq \alpha \lesssim 0.6$. However, we have results available for a greater number of alternation values in this region. For $\alpha > 0.6$, our extrapolations show deviations from the Duffy-Barr results where comparison can be made at $\alpha = 0.8$. in fact, Duffy and Barr did not attempt to extrapolate down to $T = 0$ in this alternation regime.

As part of the investigation, we have compared our spectral extrapolations with exact results for related 1D models and revealed an interesting complexity of behavior. In particular, striking differences in behavior between quantum, $S = \frac{1}{2}$, and the corresponding classical, $S = \infty$, models demonstrate the lack of validity of standard spin-wave theory, and intuition based theorem, for these quantum models.

We conclude with a final discussion of our results in relation to spin-Peierls theory and spin-exciton theory. Both theories are firmly based on the assumption of an excitation energy gap, which has recently been called into question.[30] We have established, as conclusively as seems possible in the absence of exact results, that an energy gap is present between a singlet ground state and a band of triplet-dominant low-lying excitations for all nonzero alternation, in agreement with spin-exciton theory. The nature of the excitations, whether localized or delocalized in character, could be determined by investigating the character of the corresponding finite-N eigenfunctions. The convergence of the finite-chain gap extrapolations is now expected, on theoretical grounds, to be slow near the Heisenberg uniform limit. A future attempt to improve the accuracy of the energy gap close to the uniform limit is planned.

Note added in proof. After this paper was prepared we learned that H. Matsusama and Y. Okwamoto [J. Phys. Soc. Jpn. 50, 2837 (1981)] have obtained significantly higher values for the "spin-Peierls exponents" for excitation gap and ground-state energy than those given here.

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ble limitations, of the finite-size scaling method. Finally, we express appreciation to G. Müller for a critical reading of the manuscript. This work was supported in part (for J.C.B) by the U. S. National Science Foundation under Grant No. DMR 80-10819, and by a Fellowship from the Bunting Institute, Radcliffe College, Cambridge, Massachusetts. It was also supported in part by NATO Grant No. 1338, which made possible active collaboration at the Kamerlingh Onnes Laboratorium. One of us (H.W.J.B.) acknowledges support from the research program of the Stichting voor Fundamenteel Onderzoek der Materie (FOM) which is financially supported by the Nederlandse Organisatie voor Zuiver-Wetenschappelijk Onderzoek (ZWO).

APPENDIX A: EXACT NUMERICAL CALCULATIONS

Exact machine calculations have been performed to obtain the eigenvalues of finite, alternating, antiferromagnetic Heisenberg chains of $N=2$, 4, 6, 8, 10, and 12 spins in the case of $S=\frac{1}{2}$, and $N=2$, 4, 6, and 8 in the case of $S=1$ for a variety of degrees of alternation. Use has been made of symmetry properties of the problem to block-diagonalize the original Hamiltonian matrix with dimensionality $(2S+1)^N$ such that the largest block matrix of dimension approximately 350×350 took about 20 m to solve on the University of Leiden IBM 370/158 computer system.

A similar technique was first used by Bonner and Fisher[6] (and also Griffiths[62]) to calculate the eigenvalues of uniform linear chains. Symmetry operations or conserved quantities associated with the Hamiltonian may be defined and used to create invariant subspaces of successively smaller size. These operators include the total spin S, and the Z component of the total spin $S^z=\sum_{i=1}^{N}S_i^z$. If periodic boundary conditions are used, a translation operator T may be used which corresponds to translating a given spin configuration two lattice sites around the ring. All three operators commute with the Hamiltonian H, and with each other, and therefore may be simultaneously diagonalized in an appropriate set of basis vectors $|i\rangle$. The eigenvalues are then given by

$$\vec{S}^2\,|S,S_z,T\rangle=S(S+1)\,|S,S_z,T\rangle\ ,$$

where

$$S=\frac{N}{2},\frac{N}{2}-1,\ldots,0\ ,$$

$$\vec{S}_z\,|S,S_z,T\rangle=S_z\,|S,S_z,T\rangle\ ,$$

where

$$S_z=S,S-1,\ldots,-S\ ,$$

$$\vec{T}\,|S,S_z,T\rangle=e^{2\pi ik/N}\,|S,S_z,T\rangle\ ,$$

where

$$k=0,1\cdots,\frac{N}{2}-1\ .$$

Another symmetry operator is I, which is a spin-inversion operator with eigenvalue ±1. The effect of I is to invert the Z components of the individual spins in a basis vector. Spin-inversion symmetry means that the energy eigenvalues of states for a given S^z are identical to those corresponding to $-S^z$ in zero field. A very useful operator is the (mirror) reflection operator R, which interchanges the spins at sites r and $N+1-r$. The eigenvalues of R are ±1. A systematic description of the Heisenberg Hamiltonian symmetry operators has been given.[63] In the original work of Bonner and Fisher, the symmetry operators used to reduce the Hamiltonian matrix were S^z and T. Conservation of total spin provides a very powerful way to reduce the Hamiltonian, and various approaches have since been considered which use this symmetry,[64] but the associated calculational techniques appear cumbersome and perhaps not worth the effort. A problem encountered by Bonner and Fisher was that many of the reduced submatrices contained complex elements, which increased computer running time and limited the largest N value solvable to $N=11$. The situation is that the operators in the set H, S^z, and T commute with each other, and therefore are simultaneously diagonalizable. This is also the case for the operator set H, S^z, and R. Unfortunately, the enlarged set H, S^z, T, and R cannot be simultaneously diagonalized, since R and T do not commute with each other. However, since $\vec{R}\vec{T}=\vec{T}^{-1}\vec{R}$ it is possible to combine the subspaces k and $N-k$ such that $\vec{R}$ can be diagonalized. One of us has used this property to choose an orthogonal basis set such that the associated submatrices of the Hamiltonian only contain real elements,[47,48] and further, that the Hamiltonian submatrices for the eigenvalues of T such that $k\neq0$ or $N/2$ (N even) are identical in both the $R=+1$ subspace and $R=1$ subspace. This simplifies the eigenvalue problem and is also very convenient for the calculation of eigenvectors.

APPENDIX B: EXTRAPOLATION TECHNIQUES

The limiting spectral energies as $N \to \infty$ may be estimated using the standard graphical "$1/N$" plot technique,[6] as was used to estimate the antiferromagnetic ground-state energy for uniform chains in Ref. 6. However since a large number of alternation values α were considered it was necessary to devise computer-based extrapolation techniques for convenience in handling large amounts of data. First, the gaps, G_n, obtained for finite rings of $N=2n=2$, 4, 6, 8, 10, and 12 spins were examined, and found to show a smooth behavior as a function of n, suggesting that extrapolation of the gaps to $n=\infty$ would give reliable results. Three main types of expression were used to fit the gaps, namely:

$$(1)\quad G_\infty \simeq G_n + \sum_i a_i n^{-i}\,, \tag{B1}$$

where various a_i may be put equal to zero to obtain different types of polynomials in $1/n$,

$$(2)\quad G_\infty \simeq G_n + a_i n^{-a_2}\,, \tag{B2}$$

a generalized power-law type of fit, and

$$(3)\quad G_\infty \simeq G_n + a_1 a_2^{-n}\,, \tag{B3}$$

an exponential type of fit.

A fitting procedure using Padé approximant-extrapolation techniques was also tried but was not more successful than techniques (1)–(3). Convergence rates of different types of fit were derived by comparing results for different sets of n values. Specifically, sets of three consecutive n values were chosen so that the extrapolated G_∞'s again formed a series whose convergence could be estimated. It was found that type (1) or type (2) polynomial fits gave the best results near the uniform limit ($\alpha \simeq 1$), whereas type (3) was preferable near the dimer limit ($\alpha \simeq 0$). However, in this region, say $\alpha \lesssim 0.4$, convergence was so rapid that the limit was not in doubt.

The computer-based extrapolation procedures were thoroughly tested on other systems known to show a "gap" phenomenon, vanishing at a particular limit. An example is the spin-$\frac{1}{2}$ XY alternating chain which happens to be exactly solvable for $N=\infty$, and has an alternation energy gap which vanishes in the uniform limit. These other systems are discussed in some detail in Sec. II.

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