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percent in $\langle r^2 \rangle_{av}$. We note that the experimental dispersion corrections $\Delta f'$ obtained by CJ are relatively independent of whichever theoretical model is used to calculate δf.

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Comment on Magnetic Cooling of Solid He^3 †

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The possibility of cooling solid He^3 by adiabatic magnetization is discussed using the antiferromagnetic nearest-neighbor spin-$\frac{1}{2}$ Heisenberg linear chain as a reference system. The lack of a phase transition is not important in this discussion since the locus of maximum cooling generally lies outside (i.e., is unrelated to) the T-H phase boundary, contrary to previous predictions using mean-field and spin-wave theories. Exact calculations on linear chains demonstrate a potentially useful degree of equilibrium cooling. The results are in qualitative agreement with earlier spin-wave arguments, although the details of the energy spectrum are more complicated than is assumed in spin-wave theory.

The use of adiabatic magnetization to provide a possible cooling mechanism for solid He^3 was recently discussed by Goldstein[1] on the basis of a mean-field analysis of an Ising model. Walstedt *et al.*[2] and Wolf *et al.*[3] have since independently demonstrated that no cooling is to be expected in terms of a mean-field approach because solid He^3 is an isotropic Heisenberg spin system, but that a potentially useful cooling effect is predicted using spin-wave theory. Although there is some evidence that the relaxation times may be very long,[4] the equilibrium problem is, nevertheless, of interest in its own right, and sufficiently controversial to be worthy of further discussion.

The purpose of this comment is to consider the ideas of Refs. 2 and 3 in the context of the spin-$\frac{1}{2}$ nearest-neighbor antiferromagnetic Heisenberg linear chain. At first glance the simplification to one dimension might seem to preclude any cooling, since there is no phase transition. However, a phase transition merely steepens the rate of increase of the entropy isotherms as a function of magnetic field in the region of the transition which actually occurs at a lower field than the entropy maximum, as seen in Fisher's exactly soluble model.[5] In particular, when there is a transition, the locus of maximum cooling (locus of maximum entropy) does not coincide with the T-H phase boundary, as calculated by Walstedt *et al.* using spin-wave theory, but lies outside the phase boundary in the paramagnetic region. This was first demonstrated experimentally by Schelleng and Friedberg[6] and later by Reichert *et al.*,[7] and was proved by Griffiths using a general thermodynamic argument. The general behavior of the T-H isentropes is shown schematically as Fig. 1.[8] [It may be noted that the Griffiths proof, as reported by Schelleng and Friedberg (Ref. 6), is claimed to be valid only when the specific heat C_H diverges with a critical exponent $0 < \alpha < \frac{1}{2}$ and also $\alpha = 0$ (logarithmic). However, it seems likely that this result still applies when $-1 < \alpha < 0$ (finite cusp for C_H). This range covers the expected behavior of most model antiferromagnets, both Ising and Heisenberg in type.]

The principle of the theoretical approach used by Walstedt *et al.* and by Wolf *et al.* is very simple. The thermal properties of antiferromagnets in zero field are governed by energy excitations from the ground state of the type $\epsilon(k) = \text{const} \times \sin k$, or $\epsilon(k) \simeq \text{const} \times k$ for small-k wave vectors. An applied magnetic field close to the critical value depresses the (ferromagnetic-type) levels at the top of the antiferromagnetic spectrum *below* the levels

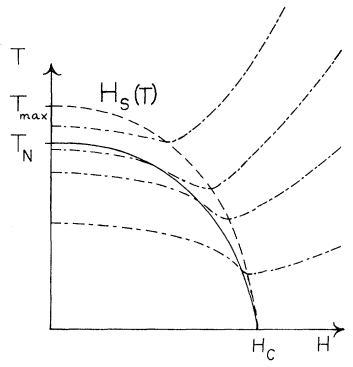

FIG. 1. Sketch of isentropes in the T-H plane for antiferromagnetic systems which show a phase transition, according to thermodynamic arguments of Griffiths. The isentropes have a point of inflection as they cross the phase boundary and are also tangent to the boundary at the crossing point. The locus of maximum cooling (dashed curve) lies outside the phase boundary (solid curve) over the whole range. However, both loci tend to $H=H_c$ as $T\to 0$.

near the antiferromagnetic ground state. The low-lying energy-dispersion relations then have the ferromagnetic form $\epsilon(k)=\text{const}\times(1-\cos k)$, or $\epsilon(k)\simeq\text{const}\times k^2$.[9] Using spin-wave theory the entropy is then estimated to behave as $S_0/N\sim aT^d$ in zero field, where d is the dimensionality of the spin lattice, and $S_c/N\sim bT^{d/2}$ at the critical field, where a and b are spin-wave constants for the particular lattice. At low enough temperatures, an adiabatic isentropic magnetization process between zero and the critical field proceeds from an initial temperature T_i to a final temperature T_f, and a measure of cooling is given by

$$(T_i-T_f)/T_f=(b/a)^{2/d}T_i^{-1}-1\simeq(b/a)^{2/d}T_i^{-1}\,, \tag{1}$$

which becomes very large as $T_i\to 0$.

We shall now test these ideas using exact calculations for finite linear chains. Our rationale for this approach is that the spin-$\frac{1}{2}$ linear chain is well known to represent the worst case as regards the applicability of spin-wave theory. If the linear chain behaves in reasonable accordance with spin-wave theory, and illustrates the predicted cooling principle, there is a strong presumption that two- and three-dimensional antiferromagnets of simple type (including the bcc phase of solid He^3) should do likewise. In Fig. 2 we show the entropy functions both along the locus of maximum entropy, $H=H_s(T)$, and in zero field, and we at once observe the features described by Walstedt *et al.* and Wolf *et al.* This figure represents an extension of earlier work by Bonner and Fisher on the behavior of the entropy as a function of field and temperature (and also of anisotropy) of one-dimensional nearest-neighbor spin-$\frac{1}{2}$ antiferromagnets.[10] Even at relatively high temperatures (where the finite-N calculations quite accurately define the large N value), where corrections to simple spin-wave theory may be expected to become important, the entropy near H_s lies above the entropy in zero field. Therefore, cooling is theoretically possible by magnetizing adiabatically from $H=0$ to $H=H_s$. [Note that the locus $H=H_s(T)$ tends to H_c as $T\to 0$.]

It is necessary to discuss the behavior in the limit as N becomes very large, however, to verify Eq. (1). For the zero-field entropy it has already been estimated that the limiting curve follows the $N=10$ curve down to temperatures of about $kT/J\simeq 0.6$, but that at lower temperatures the functional dependence on temperature is quite different.[11] The $N=10$ curve goes to zero exponentially, on account of the energy gap between the antiferromagnetic ground state and first excited states. In the limit $N\to\infty$ this gap disappears, the energy levels closing up to form a continuum, and the thermal behavior is governed by a power law in temperature. It has been estimated[11] that the entropy at low temperatures behaves as $S_0(T)/Nk\simeq 0.35\,kT/J$, where the power of T is the same as that given by spin-wave theory for one-dimensional systems. The constant, however, differs considerably from the constant $\frac{1}{3}\pi$ given by simple spin-wave theory.[12]

For the entropy near $H=H_s(T)$ a different and rather complicated pattern of convergence with increasing N is observed, which resembles quite closely the pattern of convergence for a ferromagnetic linear chain at zero field.[11] From Fig. 2 it appears that for temperatures above $kT/J\simeq 0.3$ the convergence is monotonic from below and fairly rapid, indicating a *limiting* curve lying appreciably

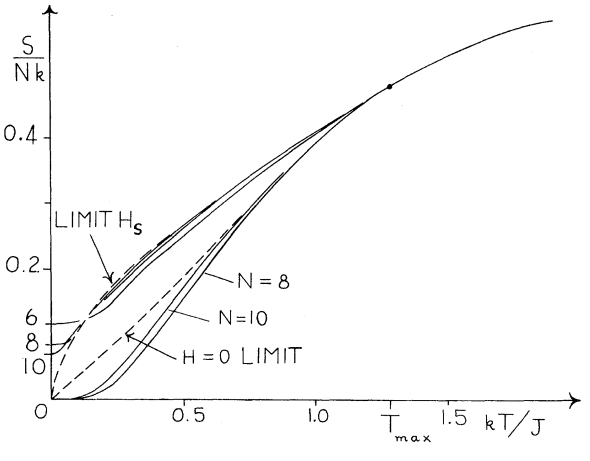

FIG. 2. Entropy curves in zero field and along the locus of maximum cooling for linear antiferromagnetic Heisenberg chains. The solid curves are exact calculations for chains of length $N=10$, 8, and 6, and the dashed curves are the corresponding $N\to\infty$ extrapolations.

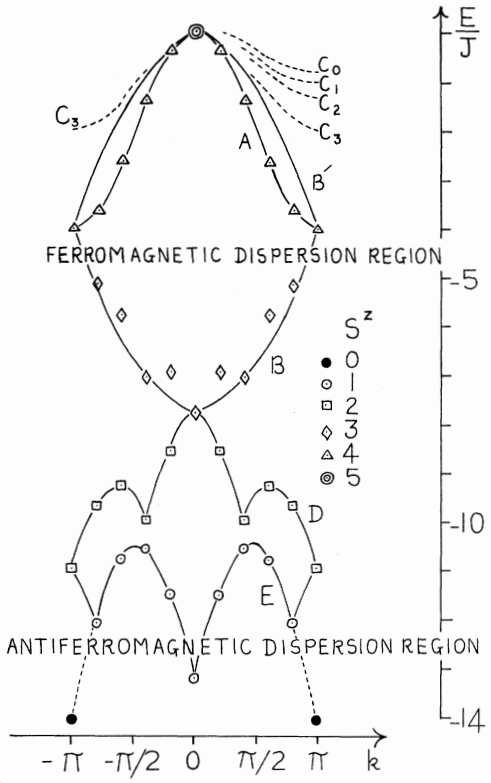

FIG. 3. Plot of important classes of energy levels for the spin-$\frac{1}{2}$ Heisenberg linear chain. The states are too numerous for all to be shown individually. The spectrum is shown for the first Brillouin zone $\pi \geq k > -\pi$. The energy levels displayed are the lowest lying for a given k and a given S^z (also for a given total spin $\vec{S}$ for $H=0$ only).

above the $H=0$ *limiting curve*, thus implying potential cooling. At lower temperatures the convergence is more complex, becoming monotonic from above very close to $T=0$, where the entropy goes to zero with increasing N as N^{-1} ln2. In the case of the ferromagnetic linear chain in zero field, the entropy has already been estimated both from purely numerical techniques and from a study of the spectrum to behave as const$\times T^{1/2}$ near $T=0$.[11] To examine whether the antiferromagnetic linear chain near the critical field shows similar behavior we now choose to study the spectrum.

In Figs. 3 and 4 we show the salient features of the spectrum of energy levels for $H=0$ and $H=H_c$, respectively, for a linear chain of **10** spins with periodic boundary conditions.[13] In **Fig.** 3 we show classes of energy states which are, in general, the lowest states in energy as a function of wave vector k for a given value of the total z component of the spin $S^z=\sum_{i=1}^{N} S_i^z$ (alternatively, of the magnetization). These classes of states lie on dispersion branches denoted by A, B, C, D, and E, where branch A corresponds to $S^z=\frac{1}{2}N-1$ $(=4)$, branch B to $S^z=3$, branch D to $S^z=2$, and branch E to $S^z=1$. Since this is an antiferromagnetic representation the (single) state corresponding to maximum S^z ($S^z=5=N/2$) is shown situated at $k=0$ at the top of the spectrum. This state is the ferromagnetic ground state. The antiferromagnetic ground state corresponds to $S^z=0$, and occurs at $k=\pi$ (or $-\pi$). The states near the antiferromagnetic ground state (curves D and E) appear to be distributed along dispersion curves which have a sin$k$-like dependence on k, in accordance with exact results of des Cloizeaux and Pearson[14] (curve E). Near the ferromagnetic limit, the curve A corresponds to a single overturned spin on the chain (single-spin-wave dispersion curve) and obeys the well-known dispersion law (energies measured from the ferromagnetic ground state) $\epsilon(k)=2J(1-\cos k)$. However, we see from Fig. 3 that other classes of states are also low lying. Curves B and B′ represent the upper and lower limits of the two-overturned-spin (two-spin-wave) continuum, obeying the dispersion relations $\epsilon(k)=4J(1\pm\cos\frac{1}{2}k)$ in the limit of large N. We observe also a special class of states, which may be identified as bound states, ly-

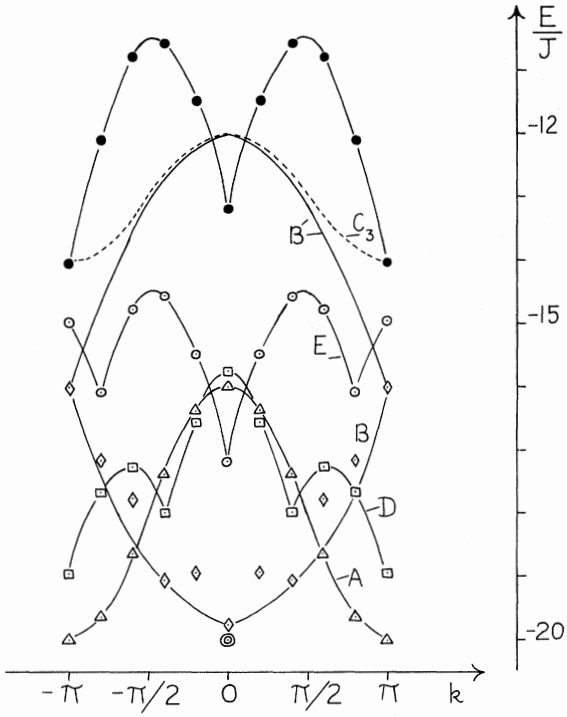

FIG. 4. Corresponding plot to Fig. 3, showing the energy-level spectrum when $H=H_c$ exactly. Whereas in Fig. 3 the complete extent of the spectrum is displayed, here we show only the lowest-lying portion. Again we show only the levels which lie lowest for a given S^z. C_3 indicates the lowest-lying bound-state dispersion curve and the only such curve within the energy range of the figure.

ing on the set of dispersion curves $\epsilon(k) = (2J/r) \times (1 - \cos k)$, where $r = 2$, 3, 4, and 5, i.e., bound spin complexes of 2, 3, 4, and 5 spins, denoted by C_i where $i = N/2 - r = S^z$. It is by no means *a priori* obvious which category of states will dominate the low-temperature ferromagnetic properties in zero field, and hence we have the well-known result that spin-wave theory is of dubious validity for the ferromagnetic linear chain.

Let us now consider the effects of an applied magnetic field[13] on the dispersion curves of Fig. 3. The sets of states will be depressed in energy by an amount proportional to their value of S^z. As H approaches the critical field at zero temperature, H_c, branches of ferromagnetic-type states from the top of the spectrum will become roughly degenerate in energy with branches of antiferromagneticlike states from the bottom of the zero-field spectrum. The situation will become more complex than in the zero-field case illustrated in Fig. 3, where it is apparent that the upper half of the spectrum is ferromagnetic in dispersion type while the lower half of the spectrum is antiferromagnetic in type. In Fig. 4 we show the low-lying portion of the whole spectrum at $H = H_c$ exactly. The ground state of the system is no longer the zero-field antiferromagnetic ground state but the former ferromagnetic ground state, which, at H_c, is degenerate with a single-spin-wave state at $k = \pm\pi$, belonging to branch A. However, the first excited state of the system is not a branch-A state but a $k = 0$ state belonging to the two-spin-wave continuum bounded by B and B′, and, generally, one- and two-spin-wave states are interlaced in energy. There is also a fairly low-lying dispersion branch D which is apparently antiferromagnetic in type: On the other hand, the families of bound states, curves C, now lie quite high up the spectrum and are relatively unimportant.

We therefore draw the following conclusions concerning the validity of spin-wave theory near H_c for the linear chain: (a) Bound states which complicate the theory of the zero-field ferromagnetic chain are not important; (b) the ground state and the lowest-lying excited states show a similar distribution to the case of the $H = 0$ ferromagnetic chain, since this distribution is materially unaffected by an inversion in energy; (c) however, antiferromagnetic-type classes of states are degenerate in energy with the one- and two-spin-wave continua, unlike the case of the zero-field chain. Despite the complication (c), it seems that the predictions of spin-wave theory near H_c should be qualitatively correct, i.e., should yield the correct functional dependence on temperature, at least for very low temperatures. At higher temperatures, where corrections to spin-wave theory may be expected to become important, the entropy calculations for finite-N systems extrapolate with increasing accuracy and predict cooling up to a temperature $kT_{\max}/J \simeq 1.28$, the temperature of the maximum in the zero-field susceptibility. Thus the cooling principle is established theoretically for antiferromagnetic linear chains. Since the linear chain is well known as the worst case for the applicability of spin-wave theory, we therefore agree with previous authors that magnetic cooling in solid He^3 may be expected if equilibrium can be achieved.

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