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Phase transitions in quasi-one-dimensional magnetic structures: Quantum effects

Y. Imry*

Department of Physics, University of California, San Diego, La Jolla, California 92037
and Department of Physics, University of California, Santa Barbara, California 93106

P. Pincus†

Department of Physics, University of California, Los Angeles, California 90024

D. Scalapino‡

Department of Physics, University of California, Santa Barbara, California 93106

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Semiclassical spin-wave theory leads to a low-temperature ($k_B T \ll J$) static correlation function for a one-dimensional Heisenberg antiferromagnet having the approximate form $\langle S_x(0) S_x(r)\rangle \propto r^{-\lambda} e^{-r/\xi}$, where $\xi(T)$ is the classical correlation length and $\lambda = 2(\pi S)^{-1}$ (S is the spin quantum number) is the quantum correction. This leads to a three-dimensional Néel temperature for exchange-coupled chains (with interchain exchange $J_\perp$), $T_N \propto (J_\perp / J)^{1/(2-\lambda)}$. The magnetic field dependence of the quantum exponent λ is shown to lead to an important field dependence for T_N.

There has been considerable recent interest in physical systems with chainlike structures which may be considered as approximately one-dimensional and thus may serve as realizations for one-dimensional theoretical models.[1] Weak coupling between the chains, which is generally present in such systems usually results in the occurrence of a three-dimensional phase transition at sufficiently low temperatures. There exist a number of magnetic systems of this type[2] which provide systematic data which may be compared with theory. We[3] have developed an approximate method for the determination of the three-dimensional (3-D) ordering temperature in weakly coupled chain systems. Our method involves two steps: (i) for isolated chains we determine the relevant one-dimensional susceptibility (uniform for a ferromagnet and staggered for an antiferromagnet); (ii) the weak interchain interactions are then treated in a molecular-field approximation to generate the 3-D phase transition. This procedure has already been carried out for some systems, i.e., the $S=\frac{1}{2}$ Ising model[4,5] and the classical Heisenberg chain[4] where the 1-D problem is exactly solvable. Both of these cases, however, are classical in the sense that all terms in the exchange Hamiltonian commute with one another. From the experimental viewpoint, quantum effects appear to play a role because the classical Heisenberg chain results agree well for systems with large spins but much more poorly for spin-$\frac{1}{2}$ systems. The purpose of this work is to investigate the effect of quantum fluctuation on T_N. We also discuss the magnetic field dependence T_N and make a speculation on the corresponding specific-heat anomaly.

The Hamiltonian for an array of weakly coupled antiferromagnetic chains has the form

$$H = 2J \sum_{\langle ij\rangle} \vec{S}_{ij} \cdot \vec{S}_{1+i,j} + 2J_\perp \sum_{\langle ij\rangle} \vec{S}_{ij} \cdot \vec{S}_{i,j+1} , \tag{1}$$

where $J \gg J_\perp > 0$. Within the framework of the approximate theory outlined above where the interchain coupling is treated as a mean field, the 3-D Néel temperature T_N is given by

$$2z J_\perp \chi(T_N) = 1 , \tag{2}$$

where χ is the staggered susceptibility of an isolated chain and z is the number of nearest neighbor chains.

In order to calculate the one-dimensional susceptibility, we consider the static correlation function,

$$G(r) = \langle S_x(0)\, S_x(r)\rangle , \tag{3}$$

where r is measured in units of the interspin separation along the chain; then the staggered susceptibility is[6]

$$\chi = (k_B T)^{-1} \int_0^\infty (-1)^r G(r)\, dr . \tag{4}$$

In the classical limit treated by Fisher and Nakamura[7]

$$G(r) \cong \tfrac{1}{3} S(S+1)\, e^{-r/\xi} (-1)^r ; \quad r \gg 1 , \tag{5}$$

where the correlation length ξ is

$$\xi = 2JS(S+1)/k_B T . \tag{6}$$

This leads to a staggered susceptibility $\chi = \frac{2}{3}(k_B T)^{-1} \times \xi S(S+1)$. Substituting this into Eq. (2) leads to the following expression for T_N:

$$(k_B T_N / J) = \frac{2}{\sqrt{3}}\, S(S+1) \left(2\, \frac{z_\perp J_\perp}{J}\right)^{1/2} . \tag{7}$$

For antiferromagnets, this theory omits the important quantum-mechanical "zero-point motion"[8] which ultimately destroys the long range ordered Néel state even at absolute zero. While we do not have an exact treatment of the quantum-spin problem, a reasonable semiclassical approximation should be given by relatively standard spin wave theory. In the vicinity of $T_N(\ll J/k_B)$, the correlation length $\xi \gg 1$ and thus for wave vectors such that $k\xi \gg 1$, the chains appear ordered and $G(r)$ (for $r<\xi$) could be calculated from spin-wave theory based on an ordered ground state. This is essentially the approach followed by Villain[9] in his "harmonic approximation" which results in

$$G(r) \cong \tfrac{1}{3}(-1)^r S(S+1)\, r^{-\lambda} e^{-r/\xi}\,, \tag{8}$$

where ξ is the classical correlation length (Eq. 6) and λ is the quantum correction. A possible unjustified extrapolation of Villain's[9] results to the isotropic Heisenberg case yields $\lambda = (\pi S)^{-1}$. A straightforward spin-wave calculation of the transverse correlation functions and including their effect on the longitudinal correlation function yields $\lambda = 2(\pi S)^{-1}$. What we wish to emphasize is that $\lambda = 0$ in the classical ($S \to \infty$) limit with an $O(S^{-1})$ correction for large S. This form correctly predicts the absence of long-range order at $T = 0°$ K for finite S. There exist several limits of this type of spin-wave approximation which provide some basis for analyzing the reliability of the results: (i) the classical limit, $S\to\infty$, is faithfully reproduced; (ii) the classical $X-Y$ model correlation function[10] is given correctly; (iii) the $S=\frac{1}{2}$ $X-Y$ model[11] at $T=0$ is predicted[9] to have the form (Eq. 8) with $\xi=\infty$ and $\lambda = \sqrt{2}/\pi \simeq 0.45$ while there exists an exact result[12] with $\lambda = 0.5$; (iv) Extrapolating these results to spin $\frac{1}{2}$ yields $\lambda = (2/\pi)$ or $(4/\pi)$, while Luther and Peschel[13] recently found $\lambda = 1$, which is also consistent with an analysis by Richards[14] of the numerical results of Bonner and Fisher.[15]

Combining Eq. (8) with Eqs. (2) and (4), we obtain the quantum corrected form for T_N

$$(k_B T_N)/J = 4S(S+1)[(zJ_\perp/3J)\,\Gamma(1-\lambda)]^{1/(2-\lambda)}\,, \tag{9}$$

where $\Gamma(x)$ is the gamma function. Comparing Eq. (7) and Eq. (9), we see that for $zJ_\perp/3J \sim 10^{-3}$, $S=\frac{1}{2}$ (boldly extrapolating the semiclassical result to the extreme quantum limit) we predict $T_N(\text{quantum})/T_N(\text{classical}) \sim 10^{-1}$, i.e., a strong further reduction of T_N arising from quantum fluctuations. The application of an external uniform field in our approximation has the striking effect of altering the exponent λ. The field-dependent correlation function may easily be calculated with this method by utilizing the spin-wave modes in a flopped (canted) antiferromagnet.[16] To lowest order in the external field H, we find

$$\lambda(H) \cong \lambda(1 - Ah^2); \quad \xi(H) \cong \xi(1 + Bh^2)\,, \tag{10}$$

where $h = g\mu_B H/4JS$ and A and B are constants of order unity. In the classical limit ($\lambda \to 0$), this essentially agrees with the recent calculations of the classical staggered susceptibility by Blume *et al.*[17] The strongest field effect on T_N arises from the reduction in λ which is effectively a suppression of the quantum fluctuations leading to a very unusual increase in T_N. For $zJ_\perp/3J \sim 10^{-3}$, $S=\frac{1}{2}$, and $h \sim \frac{1}{3}$, this leads to $T_N(h)/T_N(0) \sim 2-3$ which is semiquantitatively in agreement with the results of Azevedo *et al.*[18] in α-*bis*(N-methylsalicylaldiminato)-Cu (α-CuNSal).

The nature of the specific heat anomaly at T_N in these highly anisotropic antiferromagnets is not yet clear—neither from the experimental nor theoretical points of view. If we speculate on a second-order phase transition, then with the additional requirement of the isolated chain nonlinear staggered susceptibility[4] our mean-field approximation generates the specific heat discontinuity at T_N. Using a scaling argument[19] to determine the nonlinear susceptibility from the linear χ, we find a modified specific heat jump

$$\frac{\Delta C(\text{quantum})}{\Delta C(\text{classical})} \cong (1-\lambda/2)^2\,, \tag{11}$$

which is approximately 0.45 for $S=\frac{1}{2}$; this change is in the right direction to improve the agreement between theory and experiment.[4] This reduction in ΔC could be associated with the removal of more entropy in short-range correlations as T_N is suppressed by quantum fluctuations. Thus while the actual form[20] will differ from the mean-field prediction, we expect that the amplitude of the anomaly at T_N will be reduced by quantum fluctuations.

We have demonstrated that the strong modification of the static spin-spin correlation function by quantum-mechanical zero-point fluctuations in a quasi-one-dimensional antiferromagnet dramatically alters the three dimensional ordering temperature. In future publications we plan to present details of the calculations together with the inclusion of anisotropy field effects.

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[1]See, for example, E. H. Lieb and D. C. Mattis, *Mathematical Physics in One Dimension* (Academic, New

York, 1966).
[2]For a recent review see J. L. de Jongh and A. R. Miedema, Adv. Phys. 23, 1 (1974).
[3]D. J. Scalapino, Y. Imry, and P. Pincus, Phys. Rev. B 11, 2042 (1975).
[4]Y. Imry, D. J. Scalapino, and P. Pincus, (unpublished).
[5]J. W. Stout and R. C. Chisholm, J. Chem. Phys. 36, 979 (1962).
[6]L. Landau and E. Lifshitz, *Statistical Physics* (Pengamon, London, 1958), Chapt. XII.
[7]M. E. Fisher, Am. J. Phys. 32, 343 (1964); T. Nakamura, J. Phys. Soc. Jpn. 7, 264 (1952).
[8]For an excellent review of spin-wave theory see, F. Keffer in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1966), Vol. 18.
[9]J. Villain, J. de Phys. (Paris) 35, 27 (1974). See also H. H. Kretzen, H. J. Mikeska, and E. Patzak, Z. Phys. 271, 269 (1974).
[10]G. S. Joyce, Phys. Rev. 155, 478 (1967) and A. McGurn (unpublished).
[11]The spin-wave method is semiclassical and should give correct results in an expansion[9] in S^{-1}. The extrapolation to $S=\frac{1}{2}$ should be considered as suggestive.
[12]E. Lieb, J. Schultz, and D. Mattis, Ann. Phys. (N.Y.) 16, 406 (1961).
[13]A. Luther and I. Peschel (unpublished).
[14]P. M. Richards (private communication).
[15]J. C. Bonner and M. E. Fisher, Phys. Rev. 135, A640 (1964).
[16]T. Oguchi and F. Keffer, J. Phys. Chem. Solids 25, 405 (1964); D. Beeman and P. Pincus, Phys. Rev. 166, 359 (1968).
[17]M. Blume, P. Heller, and N. A. Lurie Phys. Rev. B 11, 4483 (1975).
[18]L. J. Azevedo, E. O. McLean, W. G. Clark, and P. F. Seligmann, submitted to the 14th Intern. Conference on Low Temperature Physics.
[19]Y. Imry (unpublished).
[20]The 3-D critical behavior may depend on the exponent λ. We would like to thank Dr. E. Siggia for a discussion on this point.