Crossover in the Heisenberg ferromagnet

1991 J. Phys.: Condens. Matter 3 4381

(http://iopscience.iop.org/0953-8984/3/24/008)

J. Phys.: Condens. Matter 3 (1991) 4381–4387. Printed in the UK

Crossover in the Heisenberg ferromagnet

R S Fishman† and G Vignale‡

† Department of Physics, SU Station, Box 5566, North Dakota State University, Fargo, ND 58105-5566, USA

‡ Department of Physics and Astronomy, University of Missouri, Columbia, MO 65211, USA

Received 24 September 1990, in final form 28 March 1991

Abstract. By comparing the free energies of the random phase approximation and of the exact expansion to order $1/z^2$, we find a crossover between a low-temperature spin-wave regime and a high-temperature non-linear regime in the magnetic state of a Heisenberg ferromagnet. Above the crossover, the many-body interactions between the spin fluctuations become important and the kinematic constraints must be maintained. This crossover is revealed by a quantum peak in the fluctuation specific heat. Experiments have observed a peak in the specific heat C/T very close to our predicted crossover temperature.

It is well-known [1] that at low temperatures, the excitations of a Heisenberg ferromagnet can be described as spin waves. The spin-wave approximation has been remarkably successful in predicting the thermodynamic and dynamic properties of a ferromagnet at low temperatures. At sufficiently high temperatures, however, the spin fluctuations become non-linear and the spin-wave description breaks down. In this paper, we calculate the crossover temperature $\bar{T}$ between the low-temperature spin-wave regime and the high-temperature non-linear regime. The crossover temperature is given by $\bar{T} \approx 0.2zJs$, where J is the exchange constant, z is the number of nearest neighbours in the lattice, and s is the spin. Because the fluctuation specific heat peaks at $\bar{T}$, the crossover can be easily observed. Since the Curie temperature T_C scales like zJs^2, the non-linear regime between $\bar{T}$ and T_C grows as the spin increases.

The breakdown of the spin-wave approximation should be expected for at least three reasons. First, the spin kinematics [2, 3] limits the number of spin deviations per site to $2s$. Because it does not restrict the number of spin deviations, the spin-wave theory violates the kinematic contraints when the density of spin waves exceeds some critical value [3]. Second, the spin-wave approximation neglects the many-body interactions [3] between spin waves, which also become important when the spin-wave density is large. Third, Wortis [3] has shown that a pair of spin waves with total momentum near the zone edge can form a bound state with energy of order zJs. The formation of bound states above the crossover temperature $\bar{T}$ implies that the spin fluctuations can no longer be described as a set of weakly interacting, particle-like excitations.

In the following calculation, we use the random phase approximation [4,5] (RPA) to represent the more general class of spin-wave theories. But our conclusions apply to any theory which yields spin-wave thermodynamics at low temperatures. To derive the crossover temperature, we compare the RPA free energy with the exact free energy expanded to order $1/z^2$. The $1/z$ expansion was originally formulated by Horwitz and Callen [6], Brout [7,8], and others [9]. The discovery of several anomalies in this expansion prevented its widespread use. Recently, Fishman and Liu [10] (FL) have established that the $1/z$ expansions of the order parameter and free energy are in fact consistent. The troubling features of the $1/z$ expansion can now be simply explained. For example, the discontinuities in the $1/z$ corrections to the free energy and entropy at the MF Curie temperature are required [10–12] for the total free energy and entropy to be continuous at the shifted Curie temperature. The other 'anomalies' can also be explained in a straightforward fashion.

Since the $1/z$ expansion was previously derived in FL, we simply sketch the method here. The Hamiltonian of the Heisenberg model is

$$H = -J \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j \tag{1}$$

where $J > 0$ is the exchange coupling and the spins obey the commutation relations

$$[S_{i\alpha}, S_{j\beta}] = -i\delta_{ij}\epsilon_{\alpha\beta\gamma}S_{i\gamma} \tag{2}$$

with $\hbar = 1$. We separate the Hamiltonian into a mean-field (MF) term H_{eff}, a constant term H_1, and a fluctuation term H_2:

$$H = H_{\mathrm{eff}} + H_1 + H_2 \tag{3}$$

$$H_{\mathrm{eff}} = -zJM_0 \sum_i S_{iz} \tag{4}$$

$$H_1 = \tfrac{1}{2}NzJ{M_0}^2 \tag{5}$$

$$H_2 = -J \sum_{\langle i,j \rangle} R_{ij} \tag{6}$$

$$R_{ij} = (S_{iz} - M_0)(S_{jz} - M_0) + S_{ix}S_{jx} + S_{iy}S_{jy} \tag{7}$$

where the MF order parameter $M_0(T^\star) = \langle S_{1z} \rangle_{\mathrm{MF}}$ is evaluated by neglecting H_2. Like every MF expectation value, M_0 is a function only of the dimensionless temperature $T^\star = T/zJ$ and of the spin s. The fluctuation term H_2 couples the local spin fluctuations on neighbouring lattice sites. As the number of nearest neighbours z increases, the mean-field experienced by each spin becomes stronger and the coupling of fluctuations becomes weaker. So the coupling of fluctuations in H_2 produces $1/z$ corrections to MF theory.

Because H_1 is constant and H_2 commutes with H_{eff}, the general expressions for the order parameter $M = \langle S_{1z} \rangle$ and partition function Z are

$$M = \frac{1}{Z}\mathrm{Tr}\left(\mathrm{e}^{-\beta H_{\mathrm{eff}}}\mathrm{e}^{-\beta H_2}S_{1z}\right) \tag{8}$$

$$Z = \mathrm{Tr}\left(\mathrm{e}^{-\beta H_{\mathrm{eff}}}\mathrm{e}^{-\beta H_2}\right) \tag{9}$$

with the free energy F defined by

$$\frac{F}{NzJ} = -T^\star \log Z + \frac{1}{2}{M_0}^2. \tag{10}$$

The $1/z$ expansion is generated by expanding equations (8) and (9) in powers of the fluctuation energy H_2. As a function of the dimensionless temperature $T^\star$, each term in this expansion can be characterized by its order in $1/z$. Formally, the order parameter and free energy may be expanded as

$$M = M_0(T^\star) + \frac{1}{z}M_1(T^\star) + \frac{1}{z^2}M_2(T^\star)\ldots \tag{11}$$

$$\frac{F}{NzJ} = \frac{F_0}{NzJ} + \frac{1}{z}\frac{F_1}{NzJ} + \frac{1}{z^2}\frac{F_2}{NzJ} + \ldots. \tag{12}$$

Both M_i and F_i/NzJ are functions only of $T^\star$ and s. Of course, the MF free energy is just F_0.

The first-order corrections M_1 and F_1 were evaluated in FL. The divergence of M_1 to $-\infty$ at the MF Curie temperature $T_C^\star = s(s+1)/3$ is not an 'anomaly' of the expansion. Rather, it signals the suppression of the Curie temperature from the MF value [10]. The first-order free energy F_1 is represented by the one-loop diagram on the bottom of figure 1. Each solid line in this diagram represents a factor of JR_{ij} which couples neighbouring lattice sites. Notice that this diagram may be positioned between any of the $Nz/2$ neighbouring sites in the lattice. So its contribution is proportional to $NzJ^2 = N(zJ)^2/z$ or, in dimensionless units, to $1/z$. The first-order fluctuation specific heat,

$$\frac{C_1(T^\star)}{Nz} = -T^\star\frac{\mathrm{d}^2}{\mathrm{d}T^{\star 2}}\frac{1}{z}\frac{F_1}{NzJ} \tag{13}$$

is plotted in the full curve of figure 2 for $s = 3/2$ and $z = 6$. As reported in FL, C_1/Nz has a peak below T_C at the temperature $\bar{T} \approx 0.2zJs$. The physical significance of this peak will be discussed shortly.

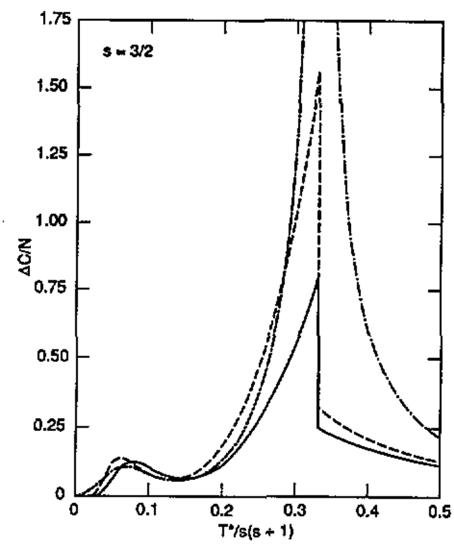

Figure 1. Diagrammatic representations of F_2/NzJ and $\Delta F^{\mathrm{RPA}}/NzJ$. F_1/NzJ is contained in the RPA ring expansion.

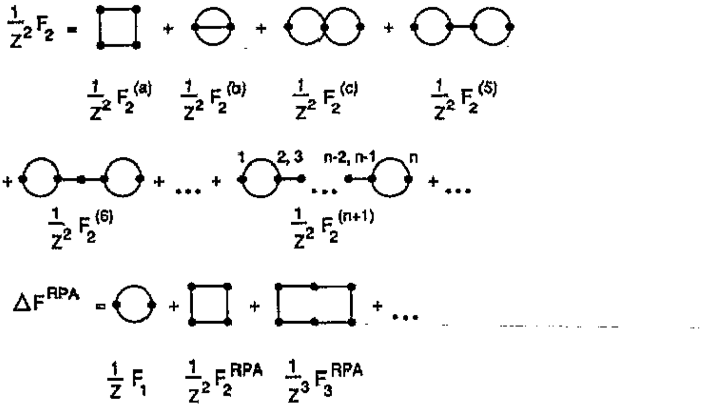

Figure 2. The fluctuation specific heat versus $T^*/s(s+1)$ for $s = 3/2$ and $z = 6$. Plotted are C_1/Nz (full), $(C_1 + C_2/z)/Nz$ (broken), and the total RPA fluctuation specific heat (chain).

It is considerably more difficult to evaluate the $1/z^2$ corrections to the free energy. The infinitely many contributions to F_2 on a d-dimensional hypercubic lattice are presented in figure 1. Most of these diagrams involve the many-body interactions between three or more fluctuations at the same site. For example, each 'dumb-bell' diagram $F_2^{(n\geqslant 5)}$ contains two sites where three spin fluctuations interact at once. Fortunately, the infinite sum over dumb-bells can be performed exactly [12]. The 'anomalous' behavior of F_2 at the MF Curie temperature is required for the continuity of the total free energy and entropy at T_C. Adding the second-order specific heat C_2/Nz^2 to C_1/Nz yields the broken curve in figure 2, where the quantum peak appears at a slightly lower temperature.

The RPA free energy F^{RPA} can also be expanded in powers of $1/z$:

$$\frac{F^{\text{RPA}}}{NzJ} = \frac{F_0}{NzJ} + \frac{1}{z}\frac{F_1}{NzJ} + \frac{1}{z^2}\frac{F_2^{\text{RPA}}}{NzJ} + \dots . \tag{14}$$

Diagramatically, the $1/z^n$ term is represented by a ring of $2n$ lines and the fluctuation free energy $\Delta F^{\text{RPA}} = F^{\text{RPA}} - F_0$ *is represented by the ring summation in figure* 1. Only two spin fluctuations interact at each vertex of a ring diagram. So unlike the exact free energy, the RPA free energy neglects the many-body interactions between the spins. The summation over rings produces the usual RPA specific heat, which in three dimensions behaves like $T^{3/2}$ near $T = 0$ and diverges at the MF Curie temperature. The ring summation also yields the RPA spin-wave frequencies

$$\omega_q = zJM_0(1 - \gamma(q)) \tag{15}$$

$$\gamma(q) = \frac{1}{z}\sum_{\delta} e^{iq\cdot\delta} \tag{16}$$

where δ are the z nearest-neighbour vectors.

The superscript RPA is omitted from the first two terms in equation (14) because they agree with the exact results of FL. So the first-order RPA free energy agrees with F_1 and the first-order RPA specific heat also peaks at $\bar{T}$. This peak survives the ring summation to appear in the *total* RPA fluctuation specific heat, plotted in the chain curve of figure 2. Since the longitudinal and transverse spin components do not couple in the ring summation of figure 1, the RPA free energy can be conveniently divided into longitudinal and transverse pieces. The peak in the fluctuation specific heat is produced by the transverse RPA free energy, $F_{\perp}^{\mathrm{RPA}}$, which contains the contributions of the spin-wave modes. As the temperature exceeds the maximum spin-wave frequency $2zJs$, spin waves with all possible momenta contribute to $F_{\perp}^{\mathrm{RPA}}$. In this equipartition regime, $F_{\perp}^{\mathrm{RPA}}$ becomes a linear function of temperature and the transverse specific heat decreases to zero, producing the peak at $\bar{T}$ [13].

To second-order in $1/z$, however, the RPA free energy differs from the exact free energy. The square diagram which represents F_2^{RPA}/NzJ is only one of the infinitely many diagrams which contribute to F_2/NzJ. Clearly, the exact free energy is far more complex than the RPA free energy. But despite their very different structures, the two free energies obey the same limit at low temperatures:

$$\lim_{T^*\to 0}\frac{F_2}{NzJ}=\lim_{T^*\to 0}\frac{F_2^{\mathrm{RPA}}}{NzJ}=-\frac{1}{8T^{*3}}s^4y \tag{17}$$

where $y=\mathrm{e}^{-s/T^*}$. Because the diagrams containing three-body interactions are proportional to y^2, they do not affect this low-temperature result. To linear order in y, the four-body interaction in $F_2^{(c)}$ can be treated as the product of two-body interactions. So to order y, only the two-body interactions are significant and F_2 agrees with F_2^{RPA}.

The difference between F_2^{RPA} and F_2 is plotted in figure 3 for three values of the spin. Notice that the free energies become significantly different only above the quantum peaks, which are indicated by arrows. So for $T<\bar{T}$, the infinite number of diagrams in F_2 can be replaced by the single square diagram of F_2^{RPA}. More generally, we believe that below $\bar{T}$ every free energy correction $F_{n\geqslant 2}$ can be replaced by the corresponding ring contribution $F_{n\geqslant 2}^{\mathrm{RPA}}$. Therefore, below $\bar{T}$ the ring summation is justified and the spin-wave description is appropriate. Above $\bar{T}$, however, the many-body interactions become important and the spin-wave approximation breaks down.

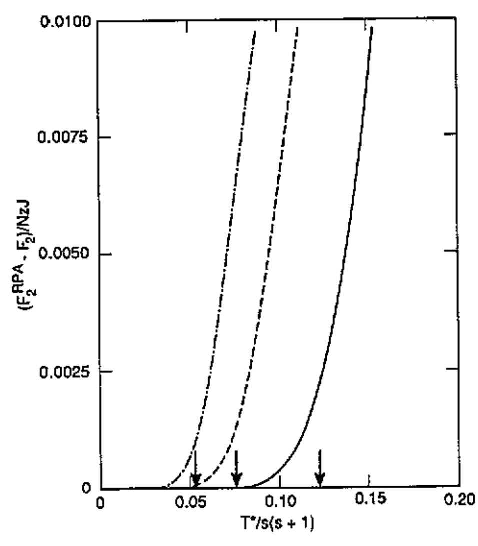

Figure 3. The difference $(F_2^{\mathrm{RPA}}-F_2)/NzJ$ versus $T^*/s(s+1)$ for $s=1/2$ (full), 3/2 (broken), and 5/2 (chain). The arrows denote the positions of the quantum peaks.

The proximity of the non-linear crossover to the quantum peak can be qualitatively understood as follows. Above $\bar{T}$, the thermodynamics is controlled by the contribution of spin waves with large momenta. As shown by Wortis [3], the interaction between two spin waves becomes stronger as their total momentum increases. So above $\bar{T}$ the spin-wave interactions must be highly non-linear. Since the spin-wave approximation cannot describe such highly non-linear interactions, it fails above the quantum peak.

Because *all* the many-body interactions contribute to the free energy above $\bar{T}$, the RPA cannot be repaired by simply adding a finite set of non-linear terms to the spin-wave Hamiltonian. For example, suppose that the three- and four-body terms are added to the spin-wave Hamiltonian. Even if the spin-wave free energy could then be evaluated *exactly*, it would still exclude the five- and six-body interactions which contribute to F_3 above $\bar{T}$. So despite a very gruelling calculation, the accuracy of that spin-wave free energy would only be of order $1/z^2$. Clearly, the easiest way to classify and examine the non-linear interactions above $\bar{T}$ is through a $1/z$ expansion.

Since the quantum peak at $\bar{T}$ appears in both the $1/z$ and RPA results for the fluctuation specific heat, this peak marks the crossover between the linear and non-linear regimes. After the large MF contribution is added to the fluctuation specific heat, the total specific heat develops a broad shoulder at the crossover temperature. Our theory predicts the shoulder to appear near $T_C/(s+1)$. Specific heat measurements on terbium [14] and gadolinium [15] have observed shoulders at temperatures very close to this prediction.

This paper disagrees with earlier workers [6–8] who supposed that every ring diagram was of order $1/z$. If that were true, the $1/z$ free energy would agree with the RPA fluctuation free energy. But for a d-dimensional hypercubic lattice, the free energy of the nth order ring diagram is proportional to

$$\frac{1}{N}\sum_{q}\gamma(q)^{2n} = \frac{1}{z^n}\frac{(2n)!}{2^n n!} + \vartheta\left(\frac{1}{z^{n+1}}\right) \tag{18}$$

not to $1/z$. So only the lowest-order ring diagram in figure 1 contributes to the first-order free energy. This example demonstrates the difference between the $1/z$ expansion and a loop expansion [16] such as the RPA, which contains terms of all orders in $1/z$.

More recently, Gros and Johnson [17] have derived the RPA from an exact expansion of the self energy to first order in $1/z$. Since the self energy enters the denominator of the spin-wave propagator, the RPA free energy is rigorous only up to order $1/z$, in agreement with this paper. In future work, we will use an expansion of the self energy to study the dynamics of the non-linear modes above $\bar{T}$.

Acknowledgments

We would like to acknowledge useful conversations with Dr C Gros, Dr M Johnson, and Dr S H Liu.

References

[1] See, for example, White R M 1983 *Quantum Theory of Magnetism* (Berlin: Springer); Mattis D C 1988 *The Theory of Magnetism I* (Berlin: Springer) chap 5
[2] Dyson F J 1956 *Phys. Rev.* **102** 1217
[3] Wortis M 1963 *Phys. Rev.* **132** 85
—— 1965 *Phys. Rev.* **138A** 1126

[4] Englert F 1960 *Phys. Rev. Lett.* **5** 102
[5] Oguchi T 1960 *Phys. Rev.* **117** 117
[6] Horwitz G and Callen H B 1961 *Phys. Rev.* **124** 1757
[7] Stinchcombe R B, Horwitz G, Englert F and Brout R 1963 *Phys. Rev.* **130** 155
[8] Brout R 1965 *Magnetism* vol 2A ed G T Rado and H Suhl (New York: Academic) p 43
[9] Vaks V G, Larkin A I and Pikin S A 1967 *Zh. Eksp. Teor. Fiz.* **53** 281 (1968 *Sov. Phys.-JETP* **26** 188)
— 1967 *Zh. Eksp. Teor. Fiz.* **53** 1089 (1968 *Sov. Phys.-JETP* **26** 647)
[10] Fishman R S and Liu S H 1989 *Phys. Rev.* B **40** 11028
[11] Fishman R S 1990 *Phys. Rev.* B **41** 4377
[12] Fishman R S and Vignale G unpublished
[13] At the crossover temperature $\bar{T} \approx 0.2zJs$, the rate of growth of the large momentum spin-wave states has reached a maximum, producing the peak in the transverse specific heat.
[14] Griffel M, Skochdople R E and Spedding F H 1954 *Phys. Rev.* **93** 657
[15] Jennings L D, Stanton R M and Spedding F H 1957 *J. Chem. Phys.* **27** 909
[16] See, for example, Negele J W and Orland H 1988 *Quantum Many-Particle Systems* (Redwood City: Addison-Wesley) chap 4
[17] Gros C and Johnson M D 1989 *Phys. Rev.* B **40** 9423