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New insight into the thermodynamics of Heisenberg ferromagnets as inferred from high-temperature series

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ABSTRACT

In search of a suitable equation of state for ferromagnets, we revise the information about the Heisenberg model obtainable from high-temperature series. Special attention is paid to the ratio χ_3/χ^4 (where χ and χ_3 are the linear and cubic susceptibilities) related to Landau's quartic coefficient b. It is found in particular that both χ_3/χ^4 and b tend to a finite limit as $T \to T_C$. This limit is small — an order of magnitude smaller than predicted by Weiss's molecular field and similar theories — but contrary to the common belief, nonzero. This implies a rejection of the generally accepted critical-point exponents and a return to those of Landau: $\alpha = 0$, $\beta = \frac{1}{2}$, $\gamma = 1$, etc.

1. Introduction

The ultimate goal of the theory of ferromagnetism is the magnetic equation of staate, $M(H, T)$. The principal difficulty here is to describe correctly the behavior of the spontaneous magnetization, $M_s(T) \equiv M(0, T)$, in the critical region, just below the Curie temperature T_C. The celebrated formula, $M_s \propto (T_C - T)^\beta$, with $\beta \approx \frac{1}{3}$, turns out to be an approximate relation. More accurate measurements on gadolinium [1] discovered no true power-law behavior at finite M_s, while the effective critical exponent β was found to tend asymptotically to $\frac{1}{2}$ as $T \to T_C$ [1]. Phenomelogically, such behavior can be described within the framework of Landau's theory [2,3]; one only needs to assume that the term in M^4 in Landau's expansion of the free energy is small and that the next term, in M^6, should be taken into account. In a previous paper [4] we proposed a simple approximate equation of state based on these ideas. The equation of state provided a good description of the $M(H, T)$ data available for elemental ferromagnets, Fe, Co, Ni, and Gd. For quantitative analysis it was proposed to present the inverse of $M_s(T)$, i.e., $T(M_s)$, as an expansion in powers of spontaneous magnetization [4],

$$\tau = 1 - a_2\sigma^2 - a_4\sigma^4 - \ldots \tag{1}$$

Here τ and σ are reduced temperature and spontaneous magnetization, $\tau = T/T_C$ and $\sigma = M_s/M_s(T = 0)$; a_2 and a_4 are coefficients *independent of* τ (as distinct from the coefficients in Landau's expansion of the free energy, who may depend on temperature). The series (1) cannot be truncated after the term in σ^2 because the 'characteristic quotient',

$$Q = \frac{a_4}{a_2}, \tag{2}$$

is large as compared with unity. Therefore, in usual experiments, where $\sigma^2 \gtrsim 0.1$, the terms in σ^2 and in σ^4 are comparable in magnitude and the dependence of σ on $1 - \tau$ does not have the form of a power law. Only in carefully controlled experimental conditions (such as in Ref. [1]) is it possible to approach the Curie point so closely that the term in σ^4 becomes negligible and Eq. (1) turns into a power-1/2 expression, $\sigma = a_2^{-1/2}(1 - \tau)^{1/2}$. The transition into this asymptotic regime takes place gradually. In most cases an approximate power law is observed, $\sigma \propto (1 - \tau)^\beta$, with β somewhere between $\frac{1}{4}$ and $\frac{1}{2}$.

Thus, it was established that Landau's theory with a large Q does agree with experiment, the large Q being crucial for the correct quantitative description of the critical behavior [4]. However, Landau's theory is a phenomenological one, it cannot predict the values of the model parameters. The search for an explanation of the large Q revealed that none of the known analytical treatments of the Heisenberg model yields a satisfactory result [4]. The molecular field approximation [5], the Oguchi approximation [5,6], the Tyablikov approximation [7,8] — they all yield $Q \approx 0.3$. That is far too little. Yet, a numerical treatment of the Heisenberg model by way of high-temperature series (HTS) expansion [9] was found to produce $Q \approx 4$ for the face-centered cubic (fcc) lattice with exchange between the nearest neighbors [4]. The Heisenberg model was thus exonerated. According to Ref. [4], it is the inadequate statistical treatment of the Heisenberg Hamiltonian in the above approximations that is responsible for the dramatic underestimation of Q. At the same time, the Kramers-Opechowski (HTS) technique was identified as the only theoretical tool capable of reproducing satisfactorily large values of Q.

Ref. [4] did not come up with a general method of mapping its Landau-type equation of state (LTES) onto the Heisenberg model. The determination of Q for the fcc lattice in Ref. [4] relied on a rare publication [9] of the $\sigma(\tau)$ dependence deduced from HTS; in Ref. [4] those data were just fitted to Eq. (1). The goal of the present work is to

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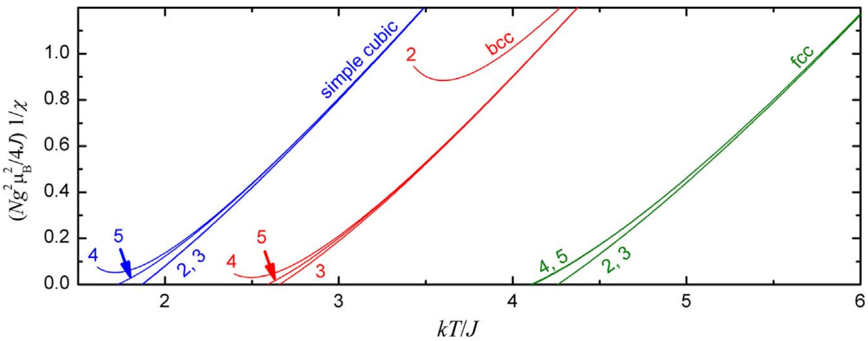

Fig. 1. Inverse susceptibility of Heisenberg ferromagnets found from HTS approximated by a diagonal Padé sequence, $[n, n]$, with $n \le 5$. Numbers on the curves are the values of n.

determine the adjustable parameters of LTES directly from HTS and other known properties of the Heisenberg model, such as the Bloch law. For simplicity we limit ourselves to spin 1/2 and cubic lattices with nearest-neighbor exchange. We do not expect that going beyond these limits will change our conclusions significantly. This paper is laid out as follows. It begins with a discussion (Section 2) of suitability of Landau's theory as a framework for describing ferromagnets near T_C, especially in connection with HTS. Such a 'second introduction' was deemed necessary because of a wide-spread opinion that HTS and Landau's theory are incompatible. Our purpose is to convince the reader that this opinion is mistaken. Section 3 is dedicated to the determination of the three model parameters of LTES by matching it with the Heisenberg model at three different temperatures. The results are discussed in Section 4. Section 5 concludes the paper.

2. Compatibility of Landau's theory with HTS

The view professed in the literature on critical phenomena [9–13] is that Landau's theory is outdated, inaccurate, valuable at best for its simplicity. The true critical behavior of ferromagnets is described by power laws with irrational exponents. Thus, paramagnetic susceptibility just above T_C depends on temperature as follows [10],

$$\chi \sim (T - T_C)^{-\gamma}, \tag{3}$$

with $\gamma \approx 1.4$. This is of course incompatible with Landau's prediction that $\gamma = 1$. So any attempt to lay the 'exact information' deduced from HTS into the procrustean bed of Landau's theory should inevitably lead to distortion of that information. Let us now see for ourselves if this view is correct.

Our starting point is the standard Heisenberg Hamiltonian,

$$\mathcal{H} = -2J\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j + g\mu_B H\sum_{i=1}^{N}S_i^z, \tag{4}$$

where $\mathbf{S}_i$ is the spin operator ($S = 1/2$) for lattice site i and the first sum is taken over all pairs of nearest neighbor sites. Following the idea proposed by Kramers and put into practice by Opechowski [14], the partition function is expanded in powers of the small parameter, $x = J/kT$. In this way one obtains, in particular, for the susceptibility at H=0 [14]:

$$\chi = \frac{Ng^2\mu_B^2}{4J}\,x\left[1 + \frac{Z}{2}x + \frac{Z(Z-2)}{4}x^2 + \dots\right] \tag{5}$$

Higher-order terms in this expansion do not depend on the coordination number Z alone, but also on a growing number of further structural parameters. Consequently, presentation of the coefficients as general explicit expressions rapidly becomes impracticable. Rather, the HTS coefficients are computed numerically for a specific lattice, by using well-established methods [15]. Thus, two decades ago Oitmaa and Bornilla [12] obtained HTS extending to terms in x^{14} (sc and bcc lattices) or x^{12} (fcc). The coefficients of the HTS present an exact algebraic result that we do not intend to dispute. It is the interpretation of the HTS that we call into question. Before presenting our arguments to the reader we would like to make the following two remarks.

(i) The series in Eq. (5) diverges near the critical temperature; so, following the common practice in the field, we shall replace the square bracket of Eq. (5) by a better-tempered diagonal sequence of Padé approximants $[n,n]$, i.e., by ratios of two polynomials of the same order n. No arbitrariness is involved in such a replacement since the coefficients of both polynomials are uniquely determined by the condition that the first $2n$ coefficients in the power expansion of the Padé approximants be the same as the coefficients in the original HTS. The available information [12] enables us to construct $[n,n]$ with $n \le 7$ (sc and bcc) or $n \le 6$ (fcc).

(ii) The susceptibility diverges as $T \to T_C$, so we choose to present graphically the inverse susceptibility, whose critical behavior is given by

$$\chi^{-1} \sim (T - T_C)^{\gamma} \tag{6}$$

The presently accepted view is that $\chi^{-1}(T)$ is non-analytical at the Curie point ($\gamma \approx 1.4$), whereas Landau [2,3] predicted a simple zero there ($\gamma = 1$).

Now the graphic evidence is presented in Figs. 1 and 2, as calculated from Padé-approximated HTS, Eq. (5), omitting the constant prefactor. Numbers on the curves are orders n of the diagonal Padé approximants to the square bracket of Eq. (5), $[n, n]$. Fig. 1 presents the early stage of research, $n \le 5$, reflecting the state of knowledge in the 1960s and 1970s. The convergence of the diagonal Padé sequence is far from being evident. The behavior of the curves near the horizontal axis in Fig. 1 vacillates between crossing the axis (a simple zero, $\gamma = 1$ in Eq. (6)) and nearly touching it, which would correspond to a double zero, $\gamma = 2$. (The latter case is characterized by a noticeably lower Curie temperature.) It was on the basis of these data that a hypothesis was put forward about the true $\chi^{-1}(T)$ being something in between: it might follow the power law (6) with γ having an intermediate value between 1 and 2. Indeed, estimates *based on this hypothesis* have consistently produced values of γ close to the geometric mean, $\gamma \approx \sqrt{2} = 1.414$. The knowledge has not evolved much over the last half-a-century: cf. $\gamma = 1.43 \pm 0.01$ [11] and $\gamma = 1.41 \pm 0.02$ [12].

Let us now turn to Fig. 2 presenting the more recent (two decades old) data [12]. One thing that draws attention is the rapid convergence of the curves with $n \ge 5$, note the scale of Fig. 2. One can hardly perceive the difference between the curves with n=6 and 7 in the left-hand panel (sc), while the corresponding curves in the middle panel are truly indistinguishable (a thousand-fold magnification would be necessary to resolve them). One is compelled to conclude that no further increase of the order of approximation n is likely to produce a distinct $\chi^{-1}(T)$ curve [16].

Secondly, the numbered lines in Fig. 2 are all but devoid of