# Critical magnetic susceptibility in the presence of long-range forces\*†

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The relatively long spin-spin force range in the ferromagnet  $GdNi_2$  renders it a prototypal system in which to compare critical exponents with calculations for a system where second-nearest-neighbor interactions are important. We have measured the low-field magnetic susceptibility of  $GdNi_2$  in the reduced temperature interval  $10^{-3} \le \epsilon \le 4$ , where  $\epsilon \equiv (T-T_C)/T_C$ . The observation of mean-field behavior over an unusually large temperature interval has allowed experimental determination of the high-temperature parameters needed for a straightforward application of the series expansions. Hence, we approximate the long-range forces by including first- and second-nearest-neighbor interactions in our Hamiltonian. High-temperature series are calculated for four different models and the experimental results are compared in detail with the classical Heisenberg and spin-infinity Ising models. The combined experimental and theoretical study suggests that universality, if it holds for this material, does so in a temperature interval sufficiently narrow about the critical point to be both experimentally and theoretically inaccessible.

#### I. INTRODUCTION

There exists considerable controversy over the hypothesis of universality1,2 of critical-point exponents at second-order phase transitions. Most of the effort has centered around the generation of high-temperature series expansions for the Heisenberg and Ising Hamiltonians. One recent study<sup>3</sup> has used indirect evidence to conclude that the critical exponents are probably independent of the strength of the second-nearest-neighbor interaction. This conclusion is contrary to numerous existing experimental results, which show a considerable variation in critical exponents.1,4 It is, of course, possible that these experimental variations arise because the measurements were not made sufficiently close to  $T_c$ . Our purpose is to provide a comparison of theory and experiment for the magnetic material GdNi2.

There are two specific difficulties which have prevented definitive comparisons between existing experiments and theories: (i) A critical exponent (say  $\gamma$ ) must be defined, in the context of data analysis, in a manner that allows for both  $T_C$  independence and the flexibility to have  $\gamma$  vary as  $T-T_C$ . (ii) There exists a number of free parameters. The parameters needed to define  $\chi$  (the susceptibility) in a high-temperature expansion, including second-nearest-neighbor interactions, are (a)  $T_C^{\rm MF}$  (the mean-field critical temperature), (b)  $\chi_0$  (the Curie constant), and (c)  $R' \equiv J_2/J_1$  (the ratio of the second- to the first-nearest-neighbor exchange constants). It will be seen that the mean-field parameters  $T_C^{\rm MF}$  and  $\chi_0$  of GdNi<sub>2</sub> are well defined from the experimental results.

## II. EXPERIMENTAL RESULTS

GdNi<sub>2</sub> is a cubic Laves-phase compound with well-localized spins on the Gd sites and no measurable moment on the Ni sites. Thus, the spin structure in GdNi<sub>2</sub> has the symmetry of the Gd sublattice, which is the diamond structure. Experimentally, we have measured the susceptibility of GdNi<sub>2</sub> with a Faraday balance in the reduced-temperature interval  $10^{-3} \le \epsilon \le 4$ , where  $\epsilon \equiv (T - T_C)/T_C$ . As  $T \to \infty$ , one expects the susceptibility to be described by mean-field theory. That is,

$$\chi^{-1} = \chi_0^{-1} (T - T_C^{MF}) \tag{2.1}$$

or

$$\frac{1}{\chi T} = \frac{1}{\chi_0} \left( 1 - \frac{T_C^{MF}}{T} \right) . \tag{2.2}$$

A search for mean-field behavior by plotting  $\chi^{-1}$ vs T, as suggested by Eq. (2.1), can often be misleading since significant changes in the behavior of the data can always be expected in the interval between the last data point and  $T = \infty$ . A more fruitful procedure is to plot  $1/\chi T$  vs 1/T, as suggested by Eq. (2.2). Thus, with  $T = \infty$  on the graph, a valid judgment can be made as to whether or not the infinite-temperature limit has been obtained. The results on GdNi2 are compared with previous work on  $EuO^5$  and  $Ni^6$  in Fig. 1. Note that only in the case of GdNi2 is there an appreciable mean-field region (straight-line behavior) from which  $\chi_0$  ( $T = \infty$  intercept) and  $T_C^{MF}$  ( $1/\chi T = 0$ intercept) can be determined. This feature of GdNi<sub>2</sub> was first observed by Cannon et al. The straight-line behavior is borne out by the agreement with the high-temperature-series curves shown in the solid and dashed lines. Also, the large region of mean-field behavior is consistent with the electrical resistivity of  $GdNi_2$ , which exhibits de Gennes-Friedel (mean-field) behavior for  $\epsilon \ge 0.1$ . As a consistency check we have obtained a value of  $7.95\mu_B$  for the Gd moment from  $\chi_0$  (the Curie constant). Within experimental error this is equal to the Gd free ion moment and is consistent with experiments suggesting that the Ni atoms do not carry a moment in this intermetallic compound.

For T sufficiently close to the critical temperature  $T_{\mathcal{C}}$  we expect

$$\chi^{-1} = A \epsilon^{\gamma} , \qquad (2.3)$$

where A is a temperature-independent constant. The critical-point exponent can then be obtained by plotting  $\chi^{-1}$  vs  $\epsilon$  as in Fig. 2. Problems arise with curves of the type shown in Fig. 2 if  $\gamma$  is allowed to vary as  $T+T_C$ . The difficulty arises because experimental uncertainty in  $T_C$  (in our case  $\delta T_C\cong\pm30$  mK) will always mask a change in  $\gamma$  sufficiently close to  $T_C$ . This is schematically shown by the dashed lines in Fig. 2. To avoid this difficulty we have chosen an approach similar to that suggested by Kouvel and Fisher. We note that if  $\chi^{-1} \propto \epsilon^{\gamma}$  with  $\gamma$  constant, then  $\ln(d\chi^{-1}/dT)$  is linear in  $\ln\chi^{-1}$ , with a slope given by  $(\gamma-1)/\gamma$ . We can then use this slope to define  $\gamma$  in the regions where  $\gamma$  is slowly varying. The exponent  $\gamma$ , defined in

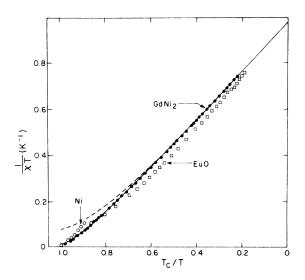


FIG. 1. Susceptibility of  $GdNi_2$ , EuO (Ref. 5), and Ni (Ref. 6). Dashed line:  $1/\chi T$  determined from the exact tenth-order series expansions for the  $S=\infty$  Ising model on a diamond lattice. Solid line:  $1/\chi T$  determined by extrapolating the exact tenth-order series to 50th order using the binomial-expansion coefficients of  $\chi = C/T(1-T_C/T)^{1/2}$  for terms 11 through 50.

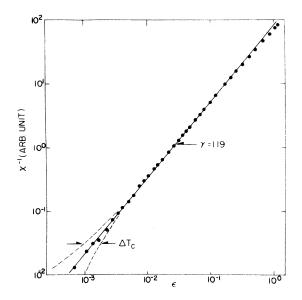


FIG. 2. Inverse susceptibility vs reduced temperature. Solid circles: experimental data on  $\mathrm{GdNi}_2$ . Dashed lines: qualitative estimate of uncertainty in  $\epsilon$  due to an uncertainty in  $T_C$  of 30 mK.

this manner, is manifestly independent of  $T_{\mathcal{C}}$ . The result of such an analysis is shown in Fig. 3. The following points are worth noting: (i) for  $\epsilon \lesssim 10^{-1}$ ,  $\gamma = 1.19 \pm 0.02$ ; (ii) as  $\chi^{-1}$  (and hence  $\epsilon$ ) grows large, the curve flattens and approaches the mean-field value of  $\gamma = 1.0$ ; and (iii) as  $T - T_C$ , there is, within experimental error, no detectable tendency for  $\gamma$  to change with temperature. Thus, Fig. 2 explicitly demonstrates the change in  $\gamma$  from a meanfield value to a "critical" one without convoluting effects from  $T_C$  renormalization. Furthermore, there is no detectable trend for  $\gamma$  to change toward a universal value as  $T - T_C$ . We have found that both this small apparent value of  $\gamma$  and the large reduced transition temperature ( $T_C/T_C^{MF} = 0.913$ ±0.012) may be consistent with the high-temperature series expansions.

### III. THEORY AND COMPARISONS

We define the interaction Hamiltonian as

$$\mathcal{H} = -\sum_{\langle i\delta \rangle} \sum_{\alpha=1}^{D} J_{\delta}^{\alpha} S_{i}^{\alpha} S_{\delta}^{\alpha} , \qquad (3.1)$$

where  $S^{\alpha}_{\delta}$  is the  $\alpha$ th component of the spin located at the  $\delta$ th site. The spin coupling parameter  $J^{\alpha}_{\delta}$  in general may be different for different  $\delta$ 's and  $\alpha$ 's. Here the summation over lattice sites  $\langle i\delta \rangle$  is not necessarily restricted to first nearest neighbors, and the sum over  $\alpha$  determines the symmetry of the spin space. For instance, if D=3 and  $J^{\alpha}_{1}=J^{\alpha}_{2}=J^{\alpha}_{2}=J^{\alpha}_{2}=J^{\alpha}_{2}$  the Hamiltonian represents

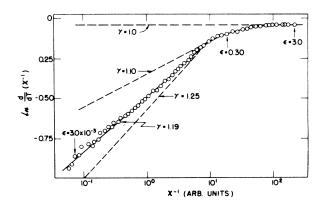


FIG. 3.  $(d/dT) \ln \chi^{-1}$  vs  $\chi^{-1}$  for GdNi<sub>2</sub>. Straight lines indicate power-law behavior (viz.,  $\chi^{-1} \alpha^{\epsilon^{\gamma}}$ ) for various values of  $\gamma$ .

the classical Heisenberg model with first- and second-nearest-neighbor interactions of equal interaction strength.

The ferromagnet  $\mathrm{GdNi}_2$  is of the cubic Laves-lattice structure with the  $\mathrm{Gd}$  atoms on the diamond lattice. Since the spin-spin interactions are believed to be of long-range nature, we will approximate the interactions by allowing the restricted sum  $\langle i\delta \rangle$  to be over first and second nearest neighbors on the diamond lattice with arbitrary interaction strengths  $J_1$  and  $J_2$ . Using a program based upon the renormalized linked-cluster theory of Wortis  $et\ al.,^{12(a)}$  we have calculated the two-spin correlation functions  $C_2(\mathbf{r})$  as high-temperature expansions in  $J_1/k_BT$  for a judiciously chosen set of values of  $R'\equiv J_2/J_1$ . From these we obtain expansions for the reduced susceptibility

$$\overline{\chi} = \frac{k_B T \chi}{N \mu^2} = \sum_{\vec{\mathbf{r}}} C_2(\vec{\mathbf{r}}) , \qquad (3.2)$$

the second moment

$$\mu_2 \equiv \sum_{\vec{\mathbf{r}}} |\vec{\mathbf{r}}|^2 C_2(\vec{\mathbf{r}}) , \qquad (3.3)$$

and the reduced specific heat 12(b)

$$\overline{C}_H \equiv \frac{TC_H}{N} = -\frac{1}{2} T \frac{\partial}{\partial T} \sum_{\vec{k}} J_6 C_2(\vec{\delta}) \tag{3.4}$$

as double power series in  $J_1/k_BT$  and R'. This was accomplished using the expansions for specific values of R' as a "basis set" and then solving simultaneous equations to obtain the general expansions. Hence the physical quantities take the form

$$\overline{\chi} = \sum_{n=0}^{\infty} \sum_{j=0}^{n} a_{nj} \left(\frac{J_1}{k_B T}\right)^n R^j$$

$$= \sum_{n=0}^{\infty} \sum_{j=0}^{n} a_{nj} \left(\frac{J_1}{k_B T}\right)^{n-j} \left(\frac{J_2}{k_B T}\right)^j . \tag{3.5}$$

We have calculated these general-R' high-temperature expansions for four different models, all on the diamond lattice. 13 (a) The first model is the traditional spin- $\frac{1}{2}$  Ising model, which is obtained by putting D=1 in Eq. (3.1) and allowing  $S=\pm \frac{1}{2}$ . Series for this model were expanded to tenth order. The second model is known as the "classical planar" model. The Hamiltonian portrays this model when D=2 and  $J_{\delta}^{x}=J_{\delta}^{y}$ , and these series were obtained to ninth order. The third model is the more familiar and perhaps more physical classical Heisenberg model. As mentioned earlier, this is obtained from the Hamiltonian (3.1) by placing D=3and  $J_{\delta}^{x} = J_{\delta}^{y} = J_{\delta}^{z}$ . The eighth-order series for  $\overline{\chi}$  and  $\overline{C}_H$  were obtained. Lastly, motivated by the unusually high experimental values of  $T_c/T_c^{MF}$ , we considered the spin-infinity Ising model. This is obtained by using the classical Heisenberg model with  $J_{\delta}^{x} = J_{\delta}^{y} \equiv 0$ . These tenth-order series can be found in NAPS. 13b Since the magnetic Gd atoms have a spin quantum number equal to  $\frac{7}{2}$  we believe the spin-infinity Hamiltonians to be a realistic approximation to GdNi2.

All the susceptibility series were analyzed by the standard-ratio, Neville, and Padé-approximant (PA) methods<sup>2</sup> for many values of R' in an attempt to find a value of R' for which both  $\gamma$  and  $T_{C}(R')/T_{C}^{MF}(R')$  agreed with experiment. In general, it was found that as R' ranged from zero to infinity,  $T_c(R')/T_c^{MF}(R')$  rose from its diamondlattice nearest-neighbor (nn) value to a broad maximum around  $R' \ge 1$ , then eventually fell back to the predicted fcc-lattice limit. Figure 4 shows this general behavior for the Ising spin-infinity and the classical Heisenberg models. The other two models gave curves whose central behavior was very similar to the Heisenberg curve and so these are not shown.14 The general behavior of the exponent  $\gamma$  also seemed to have the consistent behavior of falling off as R' increased and eventually coming back to the universal three-dimensional limits as R' reached infinity. It is interesting to note that for values of R' slightly away from the fcc limit, the Padé tables failed to show convergence. We believe this to be characteristic of the fact that for these values of R' the lattice appears to be two loosely coupled fcc lattices and the series are attempting to show certain lattice dimensionality crossover effects. 15 In Tables I(a) and I(b) of this paper we show Padé approximations to  $(\partial/\partial x) \ln \overline{\chi}(x)$ , where  $x \equiv J_1/k_B T$  for the spin- $\infty$ Ising model for R'=1 and R'=2, respectively. 16 The values of  $T_C/T_C^{\rm MF}$  and  $\gamma$  from both tables are compatible with the experimental values. The Heisenberg model [cf. Table I(c)] also showed increasing  $T_{\mathcal{C}}(R')/T_{\mathcal{C}}^{\mathrm{MF}}(R')$  and decreasing  $\gamma$ , but the  $T_c(R')/T_c^{MF}(R')$  are still much too small and the exponent too large. We believe the general

TABLE I. Estimates for the reduced inverse critical temperature  $T_C/T_C^{\rm MF}$  (upper) and the critical exponent  $\gamma$  (lower) from Padé approximates of  $d [\ln\chi(x)]/dx$  for R'=1, (a) and (c), and R'=2, (b), where  $J_1\equiv 1/k_BT$ . In this method the function is approximated by the ratio of two polynomials; N and D refer to the number of terms retained in the numerator and denominator, respectively. In (a) and (b) we present the spin-infinity Ising model results from which we estimate  $T_C/T_C^{\rm MF}=0.9074\pm0.001$ ,  $\gamma=1.185\pm0.01$  and  $T/T_C^{\rm MF}=0.9074\pm0.0005$ ,  $\gamma=1.196\pm0.01$ , respectively. In (c) we present the results for the classical Heisenberg model and we estimate that  $T_C/T_C^{\rm MF}=0.845\pm0.002$  and  $\gamma=1.31\pm0.01$ .

$D^{N}$	1	2	3	4	5	6	7	8
				(a)				
1	0.9226 1.0867	0.9166 1.1083	0.9137 1.1222	0.9119 1.1335	0.9107 1.1428	0.9098 1.1504	0.9092 1.1567	0.9088 1.1618
2	0.9266 1.0805	0.9111 1.1416	0.9086 1.1661	0.9079 $1.1752$	0.9079 $1.1763$	0.9076 $1.1792$	0.9075 $1.1814$	
3	0.9110 1.1425	0.9092 1.1595	0.9078 1.1770	0.9078 $1.1764$	0.9079 1.1746	0.9074 $1.1859$		
4	0.9087 1.1654	0.9077 1.1779	0.9079 $1.1764$	0.9078 $1.1769$	0.9071 1.1914			
5	0.9079 $1.1754$	0.9078 1.1764	0.9078 $1.1774$	0.9074 1.1856				
6	0.9079 $1.1764$	0.9079 1.1749	0.9071 1.1917					
7	0.9076 $1.1792$	0.9073 1.1863						
8	0.9075 1.1815							
				(b)				
1	0.9197 1.0881	0.9147 1.1058	0.9134 1.1126	0.9125 $1.1180$	0.9117 $1.1234$	0.9112 $1.1288$	0.9106 1.1342	0.9107 1.1393
2	0.9206 1.0864	0.9128 1.1763	0.9110 $1.1322$	0.9077 1.1889	0.9076 $1.1914$	0.9072 1.2011	0.9075 1.1916	
3	0.9127 $1.1167$	0.9117 1.1245	0.9050 1.2718	0.9076 $1.1914$	0.9077 1.1880	0.9074 1.1960		
4	0.9111 1.1314	0.9016 1.4460	0.9077 1.1883	0.9073 1.1993	0.9074 1.1961			
5	0.9072 1.2011	0.9074 1.1942	0.9072 $1.2004$	0.9074 $1.1957$				
6	0.9074 1.1941	0.9074 1.1975	0.9074 1.1970					
7	0.9072 1.2015	0.9074 1.1970						
8	0.9075 1.1915							
				(c)				
1	0.8636 1.1732	0.8586 1.1938	0.8543 1.2184	0.8511 1.2407	0.8492 1.2583	0.8479 $1.2719$		
2	0.8546 1.2215	•••	0.8432 1.3450	0.8455 1.3088	0.8454 1.3098			
3	•••	0.8414 1.3802	0.8462 1.2955	0.8454 1.3097				
4	0.8433 1.3441	0.8461 1.2970	0.8452 1.3116					

TARLE	7 T 7	(Continued)	

$D^{N}$	1	2	3	4	5	6	7	8
				(c)	***************************************			
5	0.8454 1.3091	0.8454 1.3099						
6	0.8454 1.3099							

trend of approaching the experimental values exists, and if it were possible to add third- and perhaps fourth-nn interactions one might also obtain agreement with the experimental results.<sup>17</sup>

As a further comparison of data with theory, we have plotted the *theoretical* prediction of  $1/\chi T$  vs 1/T on Fig. 1. Here we have used the spin-infinity Ising-model series with  $R' = 2.^{18}$  The dotted line corresponds to the calculated first ten terms of the expansion normalized to the experimental values of  $\chi_0$  and of  $T_{\it C}^{\it MF}$ . We should like to point out that our method lends support to the accuracy of the experimental  $T_c^{MF}$ . The normal method of curve fitting the series to the data to obtain the value of  $J_1$  is not needed, as merely scaling the first series coefficient equal to the mean-field temperature is sufficient to obtain excellent agreement with the data.20 The reader is cautioned that curves such as  $1/\chi T$  vs 1/T are relatively insensitive to R' for R' large and positive; hence Fig. 1 alone is not adequate to specify a particular value of R'.

In the above analysis we have implicitly ignored the temperature dependence of the exchange interactions which arise from thermal expansion of the lattice. In studies of the present kind, where critical exponents are studied over large temperature ranges, this effect may be formally handled by allowing the parameter  $T_{\rm C}$  to be temperature dependent. We estimate  $dT_{\rm C}/dT$  by noting that

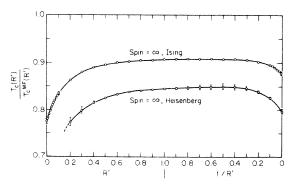


FIG. 4. Theoretical estimates for  $T_C/T_C^{MF}$  for various values of R' (= $J_2/J_1$ ) or 1/R' for the diamond lattice.

$$\frac{1}{T_C} \frac{dT_C}{dT} = \left( L \frac{\partial P}{\partial L} \right) \left( \frac{1}{L} \frac{\partial L}{\partial T} \right) \left( \frac{1}{T_C} \frac{\partial T_C}{\partial P} \right), \quad (3.6)$$

where  $L \partial P/\partial L = E$  (Young's modulus) and  $(1/L)\partial L/\partial T = \alpha$  (the coefficient of thermal expansion). Values of E typical of metals are  $(9\pm5)\times 10^{11}$  dyn/cm<sup>2</sup>. The value of  $\partial T_C/\partial P$  for GdNi<sub>2</sub> is taken as  $0.0\pm0.05$  mK/bar, 22 and from the lattice-expansion data of Zumsteg and Parks<sup>23</sup> we use  $\alpha=1\times 10^{-5}$ . Using the above results, we estimate, for GdNi<sub>2</sub>,  $dT_C/dT = (0.0\pm0.65)\times 10^{-3}$ , which gives a maximum variation in  $T_C$  of  $\pm0.2$  K over the entire experimental temperature range  $(76 \lesssim T \lesssim 350$  K). Such a small shift in  $T_c$  has a negligible effect on the results in Figs. 1–3.

## IV. DISCUSSION AND CONCLUSIONS

In summary, we have demonstrated that the experimental mean-field region has been reached, 25 yielding an accurate value of  $T_C/T_C^{MF}$ . Furthermore, the comparison of experiment and series has shown that the large experimental  $T_C/T_C^{MF}$  and low  $\gamma$  values are consistent with series predictions. The significance of this comparison is not so much in the detailed numerical agreement with the Ising model, but in the general tendency of the Padé tables to converge toward nonuniversal values of  $\gamma$ . This convergence (see below) is equivalent to a large experimental temperature region where  $\gamma$ is approximately a nonuniversal constant. This general trend also exists in the Heisenberg model. While the spin-infinity Ising model yielded critical indices comparable to experimental values, there is no a priori theoretical reason to believe the spin space is so anisotropic. In fact, experimental results for other Gd compounds indicate an isotropic Heisenberg interaction.<sup>26</sup> Hence one is left with the following question: Is the agreement with the spininfinity Ising model accidental or does the combination of Gd and Ni somehow produce and anisotropic spin space while maintaining the free magnetic moment of Gd? The second-nn classical Heisenberg model also shows critical indices moving toward the experimental values, but falling far short. Perhaps if, for the Heisenberg model, it were technically feasible to obtain lengthy series for third-(and even higher) neighbor interactions, 27 then the

experimental results could be matched equally well.

The question of whether universality has been violated can be answered on two levels. First, the apparent convergence of the Padé tables to values of  $\gamma$  similar to the experimental values would appear to suggest a possible violation. This idea is reinforced by the experimental observation of a sharp crossover from mean-field to critical behavior and by the lack of any visible temperature dependence in  $\gamma$  as  $T - T_C$  (cf. Fig. 3). However, on a second level, the following observations should be noted: (i) for R' small (i.e., short-range forces), the value of  $\gamma$  appears to approach a universal value (cf. Ref. 3). (ii) As R' gets larger [cf. Table I(a) of this paper for R'=1], the convergence of  $\gamma$  to a nonuniversal value is not good. In fact, careful scrutiny of Table I(a) reveals the suggestion of a bend toward a larger  $\gamma$  as n (number of terms in the series) increases. (iii) Given a large interspin force range [cf. Table I(b) of this paper for R'=2], the convergence of  $\gamma$  to a nonuniversal value is quite good. Experimentally this corresponds

to a large region in temperature where the critical exponent is constant and nonuniversal. This would seem to be consistent with the fact that the critical region is expected to decrease as the inverse sixth power of the force range (see, e.g., Ref. 4). Thus when the force range gets large, as in the present situation, the critical region becomes quite small and what may be left is a large region where the exponents are approximately temperature-independent nonuniversal constant. If this is indeed the experimental situation, one might ask whether or not the critical exponents in this region obey the usual scaling equalities.

Playing our own "devil's advocate," we cannot exclude the possibility that the interactions in GdNi<sub>2</sub> might not be of the general form specified by Eq. (3.1). That is, the interactions may be so long ranged that indeed one does not expect the ideas of universality to hold.<sup>28</sup>

To elucidate these questions, we are presently measuring other critical exponents since one might hope to reach the true critical region below  $T_C$ .<sup>29</sup>

<sup>\*</sup>Work forms part of Ph.D theses of two of us (P.M.H.) and (D.N.L.) to be submitted to the Physics Departments of University of Rochester and MIT, respectively.

<sup>†</sup>See NAPS document #02245 for 15 pages of Supplementary material. Order from ASIS/NAPS c/o Microfiche Publications, 305 East 46 St., New York, N. Y. 10017. Remit in advance for each NAPS accession number \$1.50 for microfiche or \$5.00 for photocopies up to 30 pages, 15¢ for each additional page. Make checks payable to Microfiche Publications.

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<sup>&</sup>lt;sup>6</sup>Data taken from P. Weiss and R. Forrer [Ann. Phys. (Paris) 5, 153 (1926)].

<sup>&</sup>lt;sup>7</sup>The susceptibility of GdNi<sub>2</sub> has been previously reported by J. A. Cannon, J. I. Budnick, M. P. Kawatra, J. A. Mydosh, and S. Kalski [Phys. Lett. A 35, 247 (1971)]. These results are qualitatively similar to the present results and, indeed, provided some of the impetus for our choosing GdNi<sub>2</sub> for the present study. The results of Cannon et al. differ in detail,

however, from the present results in that (i) Curie-Weiss behavior was claimed for  $\epsilon > 0.1$ , whereas only for  $\epsilon \gtrsim 2.0$  in the present study, and (ii) a smaller value of  $\gamma$  (viz.,  $\gamma = 1.10$ ) was obtained.

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<sup>&</sup>lt;sup>9</sup>P. G. de Gennes and J. Friedel, J. Phys. Chem. Solids 4, 71 (1958).

<sup>&</sup>lt;sup>10</sup>K. N. R. Taylor, Phys. Lett. A 29, 372 (1969). For other references see M. P. Kawatra et al., Phys. Rev. B 2, 665 (1970).

<sup>&</sup>lt;sup>11</sup>J. S. Kouvel and M. E. Fisher, Phys. Rev. 136, A1626 (1964).

<sup>&</sup>lt;sup>12</sup>(a) M. Wortis, D. Jasnow, and M. A. Moore, Phys. Rev. **185**, 805 (1969). (b) The two-spin correlation functions are defined as being in the strong interaction direction. That is, for the Heisenberg model  $C_2(r) \equiv \langle S_0^z S_r^z \rangle = \langle S_0^z S_r^z \rangle = \langle S_0^z S_r^z \rangle$ , while for the  $S = \infty$  Ising model

<sup>=</sup>  $\langle S_0^2 S_r^{x,y} \rangle = \langle S_0^2 S_r^{y,y} \rangle$ , while for the  $S = \infty$  Ising model  $C_2(r) \equiv \langle S_0^2 S_r^{x,y} \rangle \neq \langle S_0^2 S_r^{x,y} \rangle = \langle S_0^2 S_r^{y,y} \rangle$ . Using this definition it is easy to see that the reduced specific heat of Eq. (3.4) actually needs to be multiplied by the spin symmetry number given in Ref. 13(b).

<sup>&</sup>lt;sup>13</sup>(a) For the general-R series for  $\overline{\chi}$ ,  $\mu_2$ , and  $\overline{C}_H$  for all four models see NAPS document #02245 for 15 pages. Order from ASIS/NAPS. (b) We believe the accurary of these coefficients can be obtained from the formula  $N_{S.D.}$  is the number of accurate significant digits, n equals the order in inverse temperature of the coefficient, and the spin-symmetry number is defined to be equal to 1 for the Ising models, 2 for the planar model, and 3 for the Heisenberg model. Hence for the Heisenberg eighth-order coefficient  $N_{S.D.} \simeq 19 - 8 - 3 = 8$ .

<sup>&</sup>lt;sup>14</sup>For the Ising S = (1/2), planar, and Heisenberg models  $T_C(R')/T_C^{HF}(R')$  remained ≤ 0.86 for all values of R' <sup>15</sup>L. L. Liu and H. E. Stanley, Phys. Rev. Lett. **29**, 927 (1972); Phys. Rev. B **8**, 2279 (1973).

<sup>&</sup>lt;sup>16</sup>We have chosen R'=1 and R'=2 as representative points at which to compare results. These numbers by no means

should be taken to be the true experimental value of R', as series results seem to be somewhat insensitive to R' in this region.

<sup>17</sup>The number of series necessary to solve the third-nn simultaneous equation is given by (n + 2)(n + 1)/2, where n is the order of the expansion. Hence to do a tenth-order third-nn expansion one would have to have 66 different  $J_1, J_2$ ,  $J_3$  series to obtain general polynomials.

<sup>18</sup>The difference between R' = 1 and R' = 2 would be barely visible in a curve of this type.

 $^{19}J_1/k_B = T_c^{MF}/(a_{10} + a_{11}R')$ , where the  $a_{nj}$  are defined in Eq. (3.2).

<sup>20</sup>While R' is an ajustable parameter, the curves do not change much for changes in R' (see Ref. 16). This lends even more support to the idea that the experiment has reached the mean-field region.

<sup>21</sup>See, for example, K. A. Gschneidner, Jr., Rare Earth Alloys (Van Nostrand, Princeton, N.J., 1961), and D. E. Gray, American Institute of Physics Handbook (McGraw-Hill, New York, 1957).

<sup>22</sup>D. Bloch et al., J. Phys. Radium 32, C1-659 (1971); D. Chatterjee and K. N. R. Taylor, 25, 423 (1971).

<sup>23</sup>F. C. Zumsteg and R. D. Parks, J. Phys. (Paris) C1, 534 (1971).

<sup>24</sup>A scale-factor error was made in the axis labeling of Fig. 1 of Ref. 23, which may be corrected by multiplying the value of  $(1/L)(\partial L/\partial T)$  in Fig. 1 by the factor 0.40. [F. C. Zumsteg (private communication).]

<sup>25</sup>As was suggested by Cannon et al., in Ref. 7.

<sup>26</sup>J. Crangle and J. W. Ross, Proceedings of the International Conference on Magnetism, Nottingham, 1964 (The Institute of Physics and The Physical Society, London, 1965), p. 240.

<sup>27</sup>The origin of the long interspin force range in GdNi<sub>2</sub> is an interesting question in its own right. Since the Gd-Gd interaction is an indirect one, a long-range force may be generated merely be strategically placing the Gd atoms on the Ruderman-Kittel function. It is interesting to speculate, however, that while the Ni atoms carry no measurable moment, their well localized d bands may aid in the Gd-Gd information transfer through some virtual process.

<sup>28</sup>If the interspin force range decreases with distance as  $1/R^{3+\sigma}$ , where  $\sigma$  is small, one does not expect universal exponents. See, for example, M. E. Fisher, S. K. Ma, and B. G. Nickel, Phys. Rev. Lett. 29, 917 (1972).

<sup>29</sup>Below  $T_C$  the coherence length is smaller by a factor of  $\sqrt{2}$ , making the critical region larger by a factor of 8. See, for

example, Ref. 4.