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Specific heat of the classical easy-plane ferromagnetic chain with an in-plane field: A model of $CsNiF_3$

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The specific heat of the one-dimensional anisotropic classical Heisenberg ferromagnet in a magnetic field is calculated by computer simulation. The results are compared with experimental measurements on the linear-chain magnet $CsNiF_3$ and with analytical calculations on the continuum version of the model. Special attention is paid to the dependence of the specific-heat maximum on magnetic field. It is concluded that quantum effects are essential for a proper model of $CsNiF_3$.

One-dimensional magnetic systems have been the subject of intensive experimental[1,2] and theoretical studies.[3,4] Recently, Ramirez and Wolf[5] have measured the specific heat of the linear-chain magnet $CsNiF_3$ in an applied field. The magnetic excitation spectrum of $CsNiF_3$ is generally believed to be well described by the discrete spin-Hamiltonian of the ferromagnetic Heisenberg chain with single-site anisotropy[6,7]

$$\mathcal{H} = -J\sum_i \vec{S}_i\cdot\vec{S}_{i+1} + A\sum_i (S_i^x)^2 - g\mu_B H\sum_i S_i^z \ . \qquad (1)$$

For the quantum spin-1 system, the model parameters have been determined to be[1]

$$J/k_B = 23.6\ \text{K},\quad A/k_B = 9\ \text{K},\quad g\mu_B/k_B = 0.16K\ \text{kG}^{-1} \ . \qquad (2)$$

In the absence of specific-heat calculations for the model in Eq. (1), Ramirez and Wolf compare their experimental results with the theoretical predictions for the classical sine-Gordon model[8] which is taken as an approximation for Eq. (1) in the limit of extreme anisotropy,[7] $(AJ)^{1/2} >> k_B T$. They find qualitative agreement and therefore attribute the peak found in the specific heat as a function of the field to the characteristic nonlinear domain-wall excitations of the sine-Gordon model.[8] Renormalizing the domain-wall energy in a somewhat *ad hoc* manner, they even obtain a quantitative fit.

A variety of investigations,[9-11] however, have cast doubts on the validity of the sine-Gordon approximation for $CsNiF_3$, and it seems more appropriate to compare experimental results with predictions made directly for the finite-anisotropy model in Eq. (1).

In the present paper, we report the results obtained from a numerical computer-simulation study of the specific heat of the model in Eq. (1) as a function of magnetic field and temperature. We compare the numerical results with the experimental values for $CsNiF_3$ and with the low-temperature steepest-descent predictions for the continuum version of Eq. (1):

$$\mathcal{H} = \int dx\left[\tfrac{1}{2}J\left(\frac{d\vec{S}}{dx}\right)^2 + A\,(S^x)^2 - g\mu_B H(S^z-1)\right] \qquad (3)$$

recently reported by Fogedby, Hedegård, and Svane[12] and by Leung and Bishop.[13]

We find qualitative agreement but a significant quantitative discrepancy between all three sets of result. The large difference between the numerical results for the classical model in Eq. (1) and the experimental data indicates that quantum effects play a significant role.[10,11] The discrepancy between the numerical results for the discrete model in Eq. (1) and the low-temperature analytical results for its continuum version in Eq. (3) calls for an investigation of finite-temperature and finite-lattice-spacing effects.

Our computer-simulation study is based on conventional Monte Carlo importance-sampling techniques. Cyclic chains with N spins are brought to thermal equilibrium by a Glauber single-site excitation mechanism. A variety of bulk thermodynamic quantities are calculated, of which we shall here focus on the specific heat, C_H. C_H is derived via the fluctuation theorem, $C_H = (k_B T^2)^{-1}(\langle\mathcal{H}^2\rangle - \langle\mathcal{H}\rangle^2)$. The main part of the calculations is performed on chains with $N = 100$ spins. Calculations on longer chains with $N = 400$ spins show that finite-size effects can be neglected as far as C_H is concerned. It is well known that fluctuation quantities, such as C_H, are difficult to derive from computer simulations. This is particularly true of systems close to critical points. Despite the lack of true critical fluctuations in the one-dimensional spin chain studied here, it turns out to be extremely demanding to obtain accurate numerical values of C_H in the presence of the field. This is due to a very delicate competition between the ordering effect of the magnetic field term and the tendency of the entropic part of the free energy to destroy long-range ordering. Accordingly, the specific-heat results reported below are based on very extensive statistics corresponding to ensembles of about 40 000 N microconfigurations. Moreover, the final values of C_H are obtained by averaging over five to ten different ensembles constructed by using different Markov chains. Thus the statistics required for the present calculations is about two orders of magnitude larger than that needed to calculate C_H in the neighborhood of an ordinary three-dimensional critical point. We believe that similar unusual demands are responsible for the difficulties encountered by Gerling and Landau[14] in their attempt to calculate C_H for the classical XY chain in a magnetic field. In order to check the reliability of our computer simulations, we have made a comparison with the exact Fisher solution[15] for the zero-field isotropic chain $(H = A = 0)$ and with the numerically exact transfer-matrix results for the planar chain in a field $(A/J \to \infty)$.[16] In both cases, we obtain excellent agreement.

In our calculations, we have used the model parameters of Eq. (2), except for the choice of $A/k_B = 4.5$ K which is

pertinent for the classical model.[7] Selected Monte Carlo results for the magnetic field dependence of C_H at different temperatures are shown in Fig. 1. In this figure, the contribution to C_H due to the magnetic field is isolated by plotting the specific-heat difference, $\Delta C_H = C_H - C_{H=0}$. From Fig. 1, we obtain the striking result that C_H exhibits a pronounced broadened peak as the field is varied. Similar broadened peaks are observed in the specific heat of $CsNiF_3$ (Ref. 5) for which the experimental data are also included in Fig. 1. In contrast to the overall qualitative accordance between these two sets of data, there are significant quantitative differences with respect to the peak positions as well as the intensities, the latter being almost an order of magnitude smaller for $CsNiF_3$.

The low-temperature analytical calculations on the continuum model in Eq. (3) are based on the physical picture that the system at low fields forms a stable gas of spin waves and domain walls (solitons).[12,13] According to these calculations, ΔC_H is given by

$$\frac{\Delta C_H}{k_B} = \left(\frac{E_{dw}}{k_B T}\right)^2 n_s \ , \tag{4}$$

where $E_{dw} = 8(Jg\mu_B H)^{1/2}$ is the domain-wall energy and n_s is the soliton density

$$n_s = (8\pi)^{-1/2}\left(\frac{E_{dw}}{J}\right)\left(\frac{E_{dw}}{k_B T}\right)^{1/2} \mathscr{A}\left(\frac{2A}{g\mu_B H}\right) \exp\left(-\frac{E_{dw}}{k_B T}\right) \ . \tag{5}$$

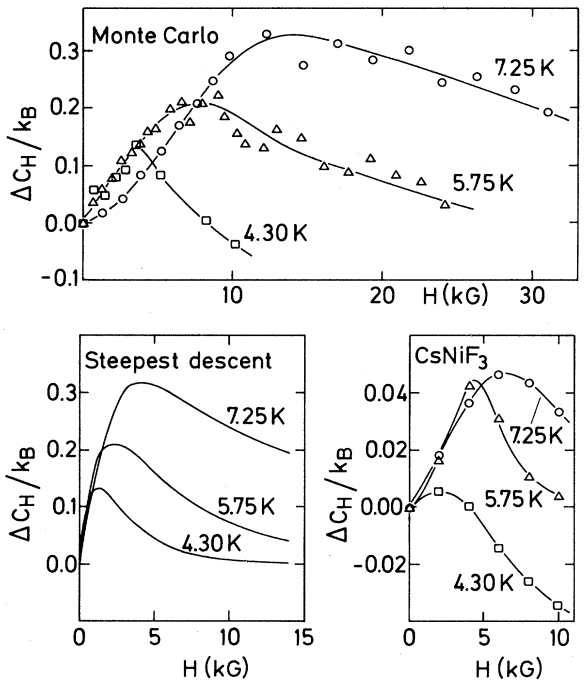

FIG. 1. Specific-heat difference $\Delta C_H = C_H - C_{H=0}$ as a function of magnetic field for three different temperatures. The experimental results of Ramirez and Wolf (Ref. 5) for $CsNiF_3$ are compared with the results of theoretical calculations on the two models in Eqs. (1) (Monte Carlo) and (3) [steepest descent, Eqs. (4)–(6)]. The scatter in the Monte Carlo data is representative of the numerical accuracy.

The anisotropy function is defined as

$$\mathscr{A}(x) = \frac{[1+(1+x)^{1/2}][2+(1+x)^{1/2}]}{(x-3)^{1/2}x^{1/2}} \ , \tag{6}$$

which reduces to unity in the sine-Gordon limit. Results derived from Eq. (4) are also shown in Fig. 1. Again, we note a qualitative accordance with the Monte Carlo results for the discrete model. Still, the peak positions are markedly different. The amplitudes, however, are very similar.

These findings strongly suggest that quantum effects in $CsNiF_3$ may be more important than hitherto expected in the temperature range considered in the experiments. This suggestion is supported by recent numerical quantum calculations on the planar model in a field[17] and on the model given in Eq. (1).[11]

The question naturally arises to which extent solitons cause the specific-heat peaks in Fig. 1. Lacking an operative and unambiguous definition of a solitary spin wave in a discrete spin chain, we are unable to deliver a definite answer to this question by the present type of Monte Carlo calculations. The analytical theories leading to Eqs. (4)–(6) relate the specific-heat maximum to the structure of the spin-wave-renormalized or "dressed" domain-wall density in Eq. (5). The validity of these theories is restricted to low temperatures, $k_B T << (Jg\mu_B H)^{1/2}$, and the question remains open whether the domain walls of the model in Eq. (3) are distinct or masked by spin-wave excitations for the range of temperatures considered here. That the specific-heat maxima are not necessarily unique features of soliton-bearing systems is perhaps most clearly indicated by the fact that we also find a broadened maximum in the Monte Carlo C_H for the isotropic ($A = 0$) chain in a field. This model is spin-wave dominated at low temperatures and does not support static finite-energy domain walls (see, e.g., Ref. 18).

The temperature dependence of the position, H_{peak}, of the specific-heat maxima in Fig. 1 is analyzed in Fig. 2 in which H_{peak} is plotted as a function of T^2. Ramirez and Wolf[5] note that their experimental data for $CsNiF_3$ are linear in such a plot, in agreement with the sine-Gordon prediction.[8] However, the experimental slope is markedly different from the sine-Gordon result (cf. Fig. 2) which can only be brought in accordance with experiments by a somewhat questionable renormalization of E_{dw}.[5] On the other hand,

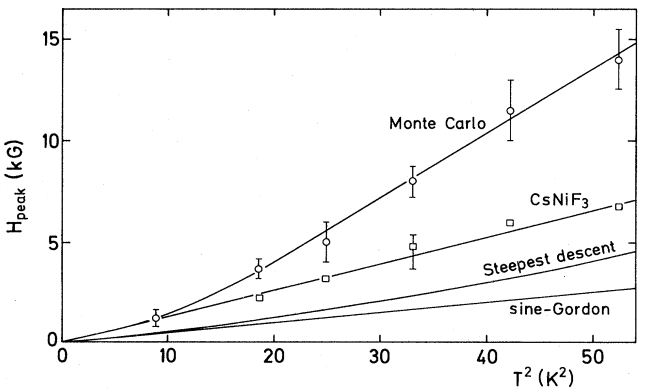

FIG. 2. Position, H_{peak}, on the field axis of the maximum of the specific heat plotted vs temperature squared. Results for three different model calculations are compared with the experimental results for $CsNiF_3$ (Ref. 5).

the analytical calculation on the finite-anisotropy continuum model in Eq. (3) (Refs. 12 and 13) gives a nonlinear dependence of H_{peak} on T^2 due to the presence of the anisotropy function in Eq. (5). The Monte Carlo results in Fig. 2 also suggest a nonlinear behavior. Since the experimental data cannot be distinguished from a linear function in the temperature interval ranging from 4 to 7 K, the nonlinear prediction can only be verified by measurements in a wider temperature range.

We wish to comment on possible explanations of the large discrepancy between the numerically exact computer-simulation results for the discrete model in Eq. (1) and the low-temperature analytical calculation on the continuum model in Eq. (3).[12,13] In order to obtain an estimate of finite-temperature corrections to H_{peak}, we have adapted the results of Sasaki and Tsuzuki[19,20] for the sine-Gordon model.[21] To order T^2, the coefficient in the linear relationship shown in Fig. 2 is found to be enhanced by a factor of $\simeq 1.7$. It is reasonable that finite-temperature corrections to the steepest-descent result for the continuum model in Eq. (3) will enhance H_{peak} with a similar factor. However, even when anticipating a correction of this size, the continuum prediction is still another factor of $\simeq 1.7$ lower than the results for the discrete model. This marked discrepancy makes worthwhile a further investigation of finite-temperature corrections as well as corrections due to the finite-lattice spacing.

Finally, a remark is in order on the anomaly predicted by the steepest-descent approach[12,13] to be present in the thermodynamic properties at a "critical" field $g\mu_B H = 2A/3$ (~ 18 kG for $CsNiF_3$). In the light of recent work[22] and in analogy with a similar behavior for the Heisenberg chain with two anisotropies,[23] this anomaly, caused by the singular structure of the anisotropy function, Eq. (6), at $x = 3$, is a spurious feature of the steepest-descent calculation which only takes into account static domain walls. At the "critical" field, the static domain wall couples via a tilting mode[12,13] to a moving domain wall, and the free energy and its derivatives (specific heat, magnetization, etc.) are expected to behave in a smooth manner. This is supported by the present work in that we do not discern such an anomaly in the Monte Carlo results, neither for the specific heat nor for any other bulk thermodynamic quantity.

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