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# Static properties of ferromagnetic quantum chains: Numerical results and experimental data on two $S = 1/2$ systems (invited)

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New results of a variant of the numerically exact quantum transfer matrix method have been compared with experimental data on the static properties of  $[\text{C}_6\text{H}_{11}\text{NH}_3]\text{CuBr}_3$  (CHAB), a ferromagnetic system with about 5% easy-plane anisotropy. Above  $T = 3.5$  K, the available data on the zero-field heat capacity, the excess heat capacity  $\Delta C = C(B) - C(B = 0)$ , and the magnetization are described with an accuracy comparable to the experimental error.

Calculations of the spin-spin correlation functions reveal that the good description of the experimental correlation length in CHAB by a classical spin model is largely accidental. The zero-field susceptibility, which can be deduced from these correlation functions, is in fair agreement with the reported experimental data between 4 and 100 K. The method also seems to yield accurate results for the chlorine isomorph, CHAC, a system with about 2% uniaxial anisotropy.

The static and dynamic properties of one-dimensional  $S = 1/2$  systems with a dominant nearest-neighbor interaction have been the subject of a large number of investigations. The Hamiltonian describing such a system is given by

$$\mathcal{H} = -2 \sum_i (J^{xx} S_i^x S_{i+1}^x + J^{yy} S_i^y S_{i+1}^y + J^{zz} S_i^z S_{i+1}^z) - \mu_B \sum_i \mathbf{S}_i \cdot \mathbf{gB}, \quad (1)$$

where  $J^{\alpha\alpha}$  ( $\alpha = x, y, z$ ) denote the components of the interaction tensor and  $\mathbf{B}$  is the external magnetic field. From a theoretical point of view, the systems described by Eq. (1) are among the most simple nontrivial model systems. Nevertheless, they display a large variety of quite unexpected features, which are not present in systems with a higher dimensionality.<sup>1</sup> These features strongly depend on the type of the anisotropy in the magnetic intrachain interaction and the magnitude and direction of the external field.<sup>2,3</sup>

Except for some special limiting cases,<sup>4,5</sup> analytical solutions of the spin Hamiltonian (1) are not available, which has stimulated many numerical studies. The oldest and probably most direct approach is the calculation of the thermodynamic properties by direct diagonalization of the Hamilton matrix for finite systems of  $N$  spins. The properties of the infinite system are then evaluated by a suitable extrapolation of the results for these finite chains.<sup>6-9</sup> In principle, this approach requires the diagonalization of a matrix with dimensions  $2^N \times 2^N$ . For systems with uniaxial symmetry, this matrix can be reduced into smaller submatrices with maximum dimensions

$$\begin{pmatrix} N \\ \frac{1}{2} N \end{pmatrix} \times \begin{pmatrix} N \\ \frac{1}{2} N \end{pmatrix},$$

which leads to a significant reduction of computer time and

memory storage. However, in the presence of exchange anisotropy or a symmetry-breaking magnetic field, such a reduction is not possible, which generally limits the chain length to  $N = 11$  or 12.

An alternative approach is based on the idea of Suzuki<sup>10</sup> to use an appropriate version of a generalized Lie-Trotter product formula to rewrite the partition function of the one-dimensional quantum system as a sum over Ising spin variables on a two-dimensional lattice. This sum has been studied for finite lattices either by classical Monte Carlo techniques<sup>11,12</sup> or by a numerically exact transfer matrix method.<sup>13</sup> The latter approach, in principle, allows the computation of the properties of chains up to a few hundred spins, thus avoiding the uncertainties introduced by extrapolation of the results for finite chains. On the other hand, results can only be obtained for finite values of the so called Trotter number  $m$ , since independent of the chain length  $N$  one has to store effectively  $2^{2m}$  numbers, representing a vector defined in a  $2m$ -dimensional Hilbert space.

The thermodynamic properties of the infinite system obtained from the various approaches mentioned above have been found to coincide at high temperatures, but at lower  $T$  significant differences are found. Of course, the accuracy of the underlying extrapolations may be estimated by comparison with the available exact results,<sup>4,5</sup> but this yields no direct information about the reliability of these extrapolations in the general case (exchange anisotropy, arbitrary orientation, or magnitude of the external field). For this purpose, a comparison with accurate data on selected experimental systems seems essential.

During the last two decades, several compounds have been synthesized that are very good realizations of theoretical model systems.<sup>14</sup> These compounds are generally built up from chains of magnetic ions that are coupled by a strong

superexchange interaction. The individual chains are effectively isolated by large organic complexes, and hence the magnetic properties of these materials above the three-dimensional ordering temperature, which is very low compared to  $J/k_B$ , resemble those of a one-dimensional system very closely.

Because of its high degree of one dimensionality and interesting magnetic properties, the compound  $[\text{C}_6\text{H}_{11}\text{NH}_3]\text{CuBr}_3$  (CHAB) has been studied very extensively. The ferromagnetic intrachain interaction in this substance contains about 5% easy-plane anisotropy.<sup>15</sup> For this type of anisotropy, the excitation spectrum may contain local nonlinear excitations, called kink solitons.<sup>16</sup> A theoretical model, in which the linear excitations were described by quantum mechanical spin-wave theory and the nonlinear excitations were associated with the solitons in the classical sine-Gordon model, was found to give a fair qualitative description of the experimental data on the heat capacity, the magnetization, the ferromagnetic resonance (FMR) spectrum, the nuclear spin-lattice relaxation time and the magnon dispersion relation.<sup>17-19</sup> A more detailed analysis, however, revealed that this nice agreement largely resulted from an accidental canceling of quantum effects by the effect of spin components out of the easy plane.<sup>20</sup> This observation also triggered direct calculations of the static properties of this system starting from the original quantum mechanical spin Hamiltonian.

For a meaningful comparison of numerical results with experimental data, the actual values of the exchange and anisotropy parameters of the compound of interest should be established as accurately as possible. With respect to the set of parameters of the spin Hamiltonian appropriate to CHAB we like to note that the anisotropies  $(J^{xx} - J^{zz})/k_B = 2.75$  K and  $(J^{xx} - J^{yy})/k_B = 0.05$  K have been determined rather directly by ferromagnetic resonance measurements<sup>17</sup> with an error that most likely does not exceed 0.02 K. The principal components of the  $g$  tensor have been determined independently from ESR experiments and measurements of the saturation magnetization. However, the experimental determination of the absolute magnitude of the intrachain interaction appeared to be less straightforward. The original analysis of the zero-field heat capacity of CHAB using extrapolated finite chain results yielded  $J^{xx}/k_B = 55 \pm 5$  K. On the other hand, a recent analysis of the magnon dispersion relation of deuterated CHAB, measured by inelastic neutron scattering,<sup>21</sup> yielded  $J^{xx}/k_B = 67 \pm 2$  K. Although we believe the latter estimate to be the most accurate, the magnitude of  $J^{xx}$  has to be considered as an "adjustable" parameter in the comparison of the available data on the static properties of CHAB with the corresponding numerical predictions. Specifically, we will try to describe both the zero-field heat capacity, the excess heat capacity  $\Delta C = C(B) - C(B=0)$ , the magnetization, and the magnetic susceptibility, using one acceptable value of  $J^{xx}$ .

In several recent studies, a variant of the quantum transfer matrix method has been applied,<sup>22,23</sup> in which the internal energy was calculated from the magnetization and all three nearest-neighbor spin correlation functions in the so called real space decomposition. In this way, a better conver-

gence in the Trotter number  $m$  has been achieved,<sup>24,25</sup> as was demonstrated by a comparison with exact results.<sup>4</sup> Actually, the internal energy was extrapolated from the results for Trotter numbers  $m = 7$  and  $m = 8$  using the  $1/m^2$  law.<sup>10,26</sup> A comparison of the results of this method with the data on CHAB revealed systematic deviations below  $T = 6$  K, which were substantially larger than the experimental uncertainties. For this reason, the original numerical method was modified as follows. First, the calculations were also performed for Trotter numbers  $m = 9$  (and partially  $m = 10$ ), and these data were included in the extrapolation. Second, the extrapolation in the Trotter number  $m$  was improved by taking into account errors in the decomposition of order  $1/m^4$ . An explicit description of the numerical transfer matrix method will be published elsewhere.<sup>27</sup>

With respect to the analysis of the thermodynamic properties of CHAB with the results of this improved method we will confine ourselves here to a rather brief description, since a detailed survey has been published very recently.<sup>28</sup> For the set of anisotropy parameters mentioned above and  $J^{xx}/k_B = 63$  K, an almost perfect agreement was found, as is illustrated in Fig. 1 for the excess heat capacity at  $B = 1, 2,$  and  $3$  T. The curves in this figure reflect the corresponding numerical results and are drawn in the region where we expect the uncertainty of the extrapolation in the Trotter number  $m$  to be smaller than 3%. It is obvious from the figure that the complete experimental behavior down to 4 K is described correctly. Small deviations of up to 5% of  $\Delta C$  seem to occur at temperatures just below the maximum for  $B = 1$  and  $2$  T. If these deviations are actually significant, they most likely result from small inaccuracies of the parameter values in the spin Hamiltonian, since in this region the uncertainty in the numerical extrapolations is much smaller.

The magnetization measured in fields up to  $B = 2$  T is plotted in Fig. 2 as  $M/M_S$  against  $B$ , where  $M_S$  denotes the saturation magnetization. The different symbols reflect sets

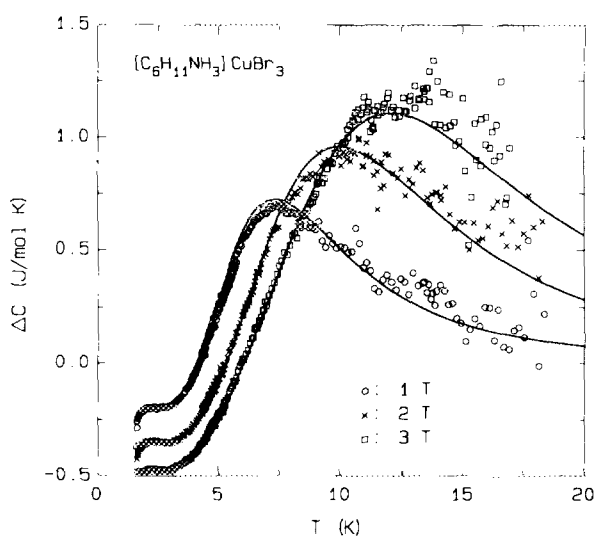


FIG. 1. Excess heat capacity  $\Delta C = C(B) - C(B=0)$  of CHAB for  $B = 1, 2,$  and  $3$  T. The curves denote the corresponding quantum transfer matrix results.

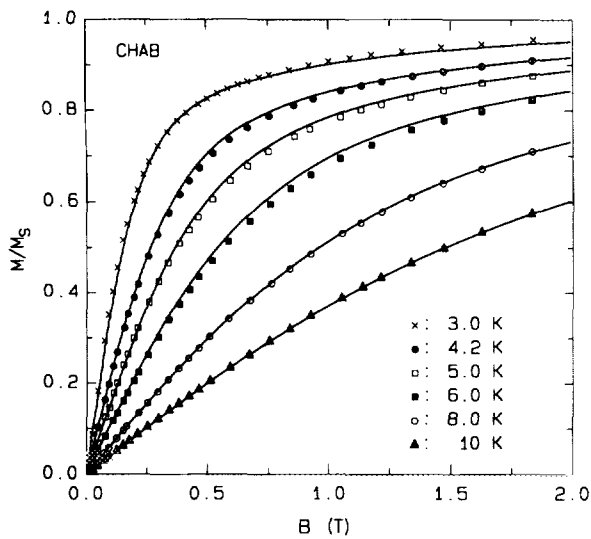


FIG. 2. Reduced magnetization of CHAB for  $\mathbf{B} \parallel \mathbf{c}$  at various temperatures. The curves denote the corresponding quantum transfer matrix results.

of measurements performed at different temperatures. The full curves reflect the corresponding behavior calculated from the modified quantum transfer matrix method for  $T \gtrsim 3$  K, where we estimate the accuracy of the numerical extrapolations to be better than a few percent. In these calculations we used the same set of parameters as in the calculation of the heat capacity. Inspection of this figure shows an almost perfect agreement between the experimental data and the numerical results; for most temperatures, the deviations do not exceed the experimental error in the determination of  $M$  (2%). Around 1 T, the data for 4.2, 5, and 6 K seem to be slightly lower than the corresponding theoretical predictions. Since in this region  $\partial M / \partial T$  is rather large, these systematic deviations should most likely be attributed to errors in the temperature measurement, because in our magnetometer the sample temperature may incidentally be 50 to 100 mK higher than that of the reference thermometer.

Since, in principle, the quantum transfer matrix method can be applied to very long chains, it is very well suited to calculations of the spin-spin correlation functions  $\langle S_i^\alpha S_{i+n}^\alpha \rangle$ ,  $\alpha = x, y, z$ , for  $n$  up to a few hundred spins. These correlation functions may serve to calculate the inverse spin-spin correlation length  $\kappa$  and the zero-field magnetic susceptibility  $\chi$  using the relations<sup>29</sup>

$$(\kappa_\alpha)^2 = 2 \lim_{N \rightarrow \infty} \left[ \sum_{n=-N}^N \langle S_0^\alpha S_n^\alpha \rangle \left( \sum_{n=-N}^N n^2 \langle S_0^\alpha S_n^\alpha \rangle \right)^{-1} \right], \quad (2)$$

$$\chi_0^{\alpha\alpha} = \lim_{N \rightarrow \infty} \frac{(g^{\alpha\alpha})^2 \mu_B^2}{k_B T} \sum_{n=-N}^N \langle S_0^\alpha S_n^\alpha \rangle. \quad (3)$$

The in-chain spin-spin correlation length in deuterated CHAB has been measured with quasi-elastic neutron scattering.<sup>30</sup> In the paramagnetic region, the correlations within the individual (decoupled) chains give rise to diffuse planes within reciprocal space perpendicular to the chain direction. The width of these planes is roughly proportional to the in-

verse correlation length  $\kappa$ .<sup>14</sup> Recently, the temperature dependence of  $\kappa_x$  for CHAB has been calculated from finite chain extrapolations and by quantum Monte Carlo techniques.<sup>12</sup> Although both the uncertainty in the experimentally deduced correlation length and the computed values of  $\kappa_x$  is rather large, the theoretical predictions for  $T > 4$  K are systematically lower than the data by a factor of 2. In order to get more information about the origin of this discrepancy, we have calculated the temperature dependence of  $\kappa_x$  using our modified quantum transfer matrix method, which we expect to be rather accurate, in view of the results presented above.

First, the correlation functions  $\langle S_0^x S_n^x \rangle$  and  $\langle S_0^z S_n^z \rangle$  were calculated for  $1 < n < 50$ ,  $2.5 < T < 50$  K and a chain of 150 spins, where  $S_0$  was located near the center of the chain. In these calculations we used the same set of parameters as for the other thermodynamic properties. Since  $S = 1/2$ , the autocorrelation functions  $\langle (S_n^\alpha)^2 \rangle$  are exactly equal to  $1/4$  for  $\alpha = x, y, z$ , all  $n$ , and all temperatures. Next,  $\kappa_x$  and  $\kappa_z$  were calculated from the two-spin correlation functions using the relation  $\langle S_0^\alpha S_n^\alpha \rangle = \langle S_0^\alpha S_{-n}^\alpha \rangle$  and Eq. (2). The correlation functions for  $n > 50$  were estimated by extrapolation of  $\langle S_0^\alpha S_n^\alpha \rangle$ , assuming exponential behavior at large  $n$ . The results are plotted as  $\kappa/T$  against  $T$  in Fig. 3, together with the reported experimental data<sup>30</sup> and the predictions from a classical spin model<sup>31</sup> with  $\tilde{S} = 1/2$ , calculated for the same set of exchange and anisotropy parameters. Inspection of this figure reveals large discrepancies between the quantum transfer matrix results and the data. In Ref. 28 it is pointed out that these deviations should be attributed to oversimplifications in the analysis of the observed neutron scattering profile. On the other hand, the quantum transfer matrix results indicate that the "cross over" between isotropic (Hei-

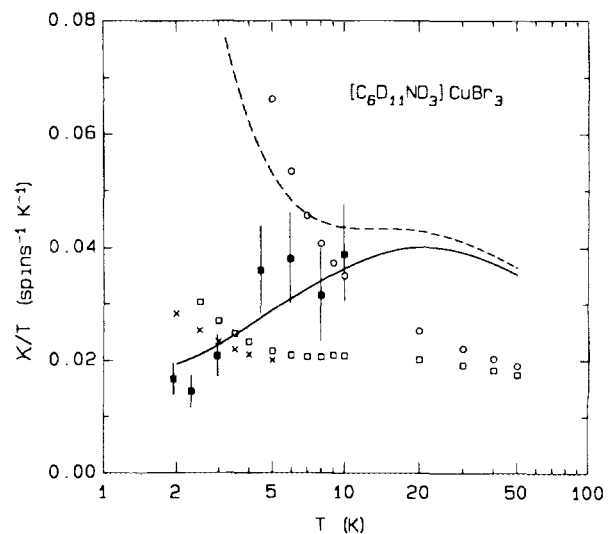


FIG. 3. Inverse correlation length of CHAB plotted as  $\kappa/T$  against  $T$ . Black squares represent the reported neutron scattering results (Ref. 30). The solid and dashed curves represent the classical spin predictions for the in-plane and out-of-plane spin components, respectively. The quantum transfer matrix results for the in-plane and out-of-plane spin components are denoted by open squares and circles, respectively. In all these calculations the small in-plane anisotropy ( $J^{xx} - J^{yy}$ ) has been neglected. The crosses reflect the effect of an appropriate in-plane anisotropy.

senberg) behavior at high  $T$  to anisotropic ( $XY$ -like) behavior at low  $T$  occurs at much higher temperatures than suggested by the classical spin model. Hence the nice description of the experimental data by the predictions for  $\kappa^x$  of this latter model should be considered as largely accidental, especially since in the experimental neutron scattering configuration both the  $x$  and  $z$  spin components are measured simultaneously.

We will conclude the present comparison of the experimentally determined static properties of CHAB with quantum transfer matrix results by noting that the same set of correlation functions that we used to calculate the inverse correlation length according to Eq. (2) were also used to calculate the zero-field magnetic susceptibility according to Eq. (3). The results were found to describe the corresponding experimental data<sup>32</sup> between 4 and 100 K fairly well, corroborating the correctness of the parameters in the spin Hamiltonian (1) and the reliability of the present numerical approach. An extension of the calculations to temperatures below 3 K would probably require an increase of the chain length  $N$ , but, more definitely, an increase of the maximum Trotter number  $m$ . In this respect we note that calculations of  $\langle S_0^x S_n^x \rangle$  for a chain of 500 spins and  $T = 3$  K yielded results that are, up to  $n = 50$ , equal to those of similar calculations for  $N = 150$ . This demonstrates again that the present numerical results are hardly affected by the finite length of the chain.

A second almost ideal realization of a ferromagnetic  $S = 1/2$  chain system is the chlorine isomorph of CHAB [ $C_6H_{11}NH_3$ ]CuCl<sub>3</sub> (CHAC). The intrachain interaction in this compound is of the same order of magnitude as that in CHAB, but, instead of an easy-plane character, the anisotropy in CHAC has an orthorhombic or XYZ symmetry.<sup>17</sup> In such a system, the excitation spectrum is predicted to consist mainly of (linear) magnons and bound complexes of two or more magnons, so-called magnon bound states.<sup>2</sup> Some experimental evidence for the presence of these magnon bound states in CHAC has been obtained from FMR measurements.<sup>33</sup> A recent analysis of the differential magnetic susceptibility of this compound indicates that also the thermodynamic properties are dominated by these excitations in a certain range of fields and temperatures.<sup>34</sup> A quantitative agreement between theory and experiment is, however, still lacking. It is not clear whether this is due to inadequacies in the theory or to uncertainties in the values of one or more parameters in the spin Hamiltonian. In this respect we like to note that also for CHAC accurate estimates of the anisotropy parameters  $(J^{xx} - J^{zz})/k_B = 1.03$  K,  $(J^{xx} - J^{yy})/k_B = 0.53$  K have been deduced from FMR experiments,<sup>17</sup> whereas the principal components of the  $g$  tensor have been determined directly from ESR experiments.<sup>35</sup> However, the reported values for the absolute magnitude of the intrachain interaction show large deviations. An analysis of the zero-field heat capacity with extrapolated finite chain results, assuming perfect uniaxial symmetry, yielded  $J^{xx}/k_B = 45 \pm 5$  K.<sup>15</sup> On the other hand, from fits of the zero field susceptibility with the results of high-temperature series expansions a value  $J^{xx}/k_B = 70$  K was inferred.<sup>35</sup>

Attempts to describe the thermodynamic properties of

CHAC by extrapolated results of the numerical diagonalization of the Hamilton matrix for finite chains using the complete spin Hamiltonian were not very successful. Especially at low temperatures, the differences between extrapolations based on a maximum chain length  $N = 10$  or  $N = 11$ , respectively, were found to be very large, which may be due to the large orthorhombic distortion of the Heisenberg symmetry.

We expect that a better description of the available experimental data can be obtained from calculations based on the modified quantum transfer matrix method, which yielded a very good agreement for CHAB. A first comparison of the experimental zero-field susceptibility of CHAC along the crystallographic  $c$  axis with our numerical results suggests that the value of  $J^{xx}/k_B$  is close to 60 K. To verify this result, we will try to obtain a consistent description of the various thermodynamic properties of this compound. The relevant experimental data include: the zero-field susceptibility along all three crystallographic axes, the powder susceptibility in applied magnetic fields, the zero-field heat capacity, and the magnetization and heat capacity for fields near the "easy" ( $x$ ) and along the "intermediate" ( $y$ ) anisotropy axis. An analysis of these data will provide rather accurate estimates of the intrachain interaction parameters in CHAC, allowing reliable checks of the validity of various theoretical predictions for the excitation spectrum in  $XYZ$  systems. Apart from this, such an analysis may serve to test the applicability of the modified quantum transfer matrix method to systems with an orthorhombic symmetry.

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