Finite-chain approach to the study of $CsNiCl_3$

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The specific heat and magnetic susceptibility of the antiferromagnetic Heisenberg chain have been calculated using a numerical finite-size technique. An agreement with experiment for the quasi-one-dimensional compound $CsNiCl_3$, which displays a Haldane gap, is revealed.

In some recent papers[1-3] we have applied a numerical finite-size method to study the thermodynamic properties of two compounds, CHAB $[(C_6H_{11}NH_3)\ CuBr_3]$ and CHAC $[(C_6H_{11}NH_3)\ CuCl_3]$, which are well represented by spin $-\frac{1}{2}$ ferromagnetic chains. In this paper we extend previous finite-size results to spin-1 systems. We focus our attention on $CsNiCl_3$, a good realization of a quasi-one-dimensional antiferromagnetic system,[4] which has recently been employed to yield experimental evidence of the Haldan gap.[5,6]

Quasi-one-dimensional magnets have attracted a great deal of interest due to nonlinear spin dynamic effects[7] and the recognition of the role of the spin value for the ground-state critical properties.[8] For $CsNiCl_3$, it has been established from susceptibility measurements[4] $J/k_B=26$ K and $g=2.20$, in agreement with the specific-heat measurements,[9] yielding $J/k_B=27$ K. However, recent neutron-scattering experiments,[5,6] performed in order to test the Haldane conjecture, have led to a somewhat higher value, $J/k_B=33.2$ K. Since the experimental temperature range $10\text{K}\leq T\leq 60$ K is accessible by our numerical calculations, we have performed a complete fitting procedure, taking into account the experimental data for the specific heat and for both the parallel ($\chi_\parallel$) and perpendicular ($\chi_\perp$) susceptibility.

The finite-size numerical approach follows the lines of our previous papers.[2,10,11] We consider a finite chain of N spins, with $N\leq 8$, described by the Hamiltonian

$$H=\sum_i \mathbf{S}_i\cdot\mathbf{S}_{i+1}+D\sum_i (S_i^z)^2 \tag{1}$$

and we impose free-boundary conditions. We diagonalize the Hamiltonian, taking advantage of the conservation of the z component of the total spin when the external field is zero or is applied along the chain. We also use the following symmetries: reversal of all the spins in the z direction, and inversion of the chain with respect to the center. We express the specific heat and the susceptibility in terms of the eigenvalues and eigenvectors for a chain of N spins,[10] and we perform a linear extrapolation for $1/N\rightarrow 0$.

From our finite-size calculations, we have found the best fit with the following set of parameters:

$$J/k_B=27\pm 3\ \text{K},\quad D/J=-0.05,\quad g=2.23\ . \tag{2}$$

Our values confirm the previous findings. In particular, our g value agrees very well with the one found from the ESR experiment.[12] The slight anisotropy in the experi-

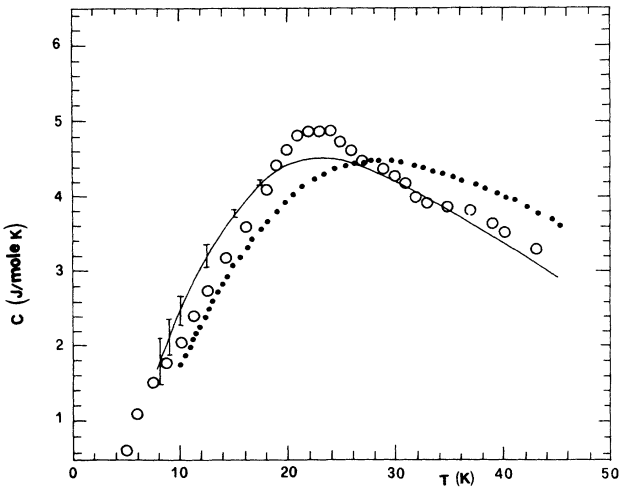

FIG. 1. Specific heat of $CsNiCl_3$. The open circles refer to experimental data. The solid line with error bars gives the finite-chain results for $J/k_B=27$ K and $D/J=-0.05$. The dotted line refers to $J/k_B=33.2$ K.

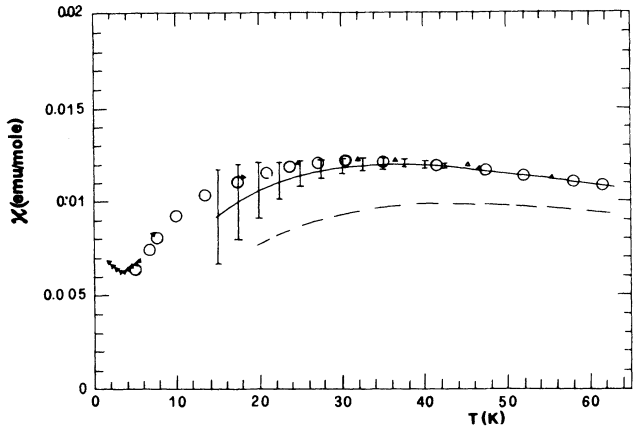

FIG. 2. Susceptibility of $CsNiCl_3$. Solid line: numerical data for $J/k_B=27$ K and $D/J=-0.05$. Dashed line: our data for $J/k_B=33.2$ K. Errors bars are shown for the solid line. The symbols report the experimental data: the circles $\chi_\parallel$ and the triangles $\chi_\perp$.

mental susceptibility data can be accounted for by a small anisotropy in g [12].

Our numerical results for the zero-field specific heat are represented in Fig. 1 as the solid line, whereas the dotted line gives the numerical results for $J/k_B = 33.2$ K. The corresponding experimental data are represented by the circles.

In Fig. 2 we plot our numerical predictions for the parallel susceptibility. The symbols represent the experimental data: the open circles refer to the parallel suceptibility and the triangles to the perpendicular susceptibility. To avoid an overlap, we skip our estimates for $\chi_\perp$. The dashed curve represents the results we find for $\chi_\parallel$ using the value $J/k_B = 33.2$ K; they underestimate the experimental data and the deviations are well above the uncertainties in our extrapolations, denoted in Fig. 2 by the vertical error bars.

In conclusion, we have successfully applied our numerical approach to $CsNiCl_3$. We have revealed a quantitative agreement with experiment over a wide temperature range for the zero-field specific heat and for the parallel and perpendicular susceptibility. We have also determined that the value of the exchange parameter inferred from the neutron-scattering experiment, performed to test the Haldane conjecture, is not consistent with the static measurements carried out to date.

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