Dispersion of magnetic excitations in a spin-1 chain with easy-plane anisotropy

O. Golinelli, Th. Jolicoeur, and R. Lacaze
Service de Physique Théorique, Centre d'Etudes Nucléaires de Saclay, F-91191 Gif-sur-Yvette CEDEX, France
(Received 9 March 1992)

We study the dispersion of magnetic excitations in a Heisenberg antiferromagnetic chain of spin $S=1$ with exchange J and single-ion anisotropy D. Chains of length up to 18 sites are studied by a Lanczös method. For the Haldane phase $D<D_c\approx J$ we show that the gap at zero momentum and $S^z=1$ is given by the sum of the gaps in the sectors $S^z=1$ and $S^z=0$ at momentum π. The transition region near $D=J$ is likely to be reduced to an isolated point. Large-D perturbation theory is smoothly recovered as soon as $D>2J$. We discuss the implications for experiments on $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP) as well as $CsFeBr_3$.

Antiferromagnetic (AF) quantum spin chains have been the subject of intense theoretical and experimental studies since Haldane's conjecture[1,2] about the difference between integer and half-integer spins. Integer spin chains are predicted to be generically massive. In the case of the isotropic Heisenberg $S=1$ chain with nearest-neighbor exchange, there is now convincing evidence from numerical studies[3-5] that a finite gap exists in the thermodynamic limit. On a finite chain the lowest-lying levels are a singlet ground state with zero momentum and a degenerate triplet $S=1$ with momentum π. The gap between these low-lying levels converges towards a finite nonzero value in the thermodynamic limit. Current estimates[3-5] for this so-called Haldane gap are close to 0.41 J. However, most studies have been restricted to the $k=\pi$ part of the spectrum of elementary excitations. In the isotropic case, a quantum Monte Carlo[5] study has given weight to the belief that the gap at $k=0$ is twice the gap at $k=\pi$ as occurs naturally in the nonlinear σ model picture of the spin-1 chain.[2]

In this paper, we obtain the spectrum of elementary excitations for a spin-1 chain with easy-plane single-ion anisotropy D. When D is increased, the Haldane gap is diminished until at $D\approx J$ it vanishes. At this point a transition[6] occurs so that when D is further increased we observe the rise of a gap of a different nature in a singlet phase.[7] These results are relevant to the interpretation of experiments on $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP),[8] for example, on the Haldane side and on $CsFeBr_3$ on the strongly anisotropic side.[9]

Experimentally the compound $Ni(C_2H_8N_2)_2NO_2ClO_4$ is one of the best candidates[10] to display the Haldane gap. The nickel ions have spin 1 and are tentatively described by the following Hamiltonian:

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

The vectors $\mathbf{S}_i$ are quantum spin operators satisfying the SU(2) rotation algebra with length $\mathbf{S}_i^2=2$. They are located along a one-dimensional lattice of N sites with periodic boundary conditions and the exchange constant J is positive. In the case of NENP, present estimates[11,12] are $J/k_B=43.5$ K and $D/J=0.18$. The isotropic chain $D=0$ can be described[1,2] by a continuum field theory, the O(3) nonlinear σ model. This is a massive theory as seen from general renormalization-group arguments as well as from the Bethe-ansatz solution. Moreover, its scattering S matrix is known and there are no bound states: the spectrum consists of a massive triplet. This has important consequences for the spin chain: the excitation near wave vector $k=0$ should consist of two massive particles and thus the gap in the $k=0$ sector is predicted to be twice the gap in the $k=\pi$ sector where the fundamental massive particle shows up. This property is thus part of the Haldane conjecture and it is worth testing it on the experimental candidates such as NENP.[13]

In the presence of single-ion anisotropy, the Haldane gap is split into two components: the triplet with $S=1$ and $k=\pi$ under the perturbation caused by the D term gives a high-energy singlet state with $S^z=0$ and a low-lying doublet $S^z=\pm1$. The gap between the ground state and the doublet (singlet) will hereafter be called $G^{(-)}$ ($G^{(+)}$). The evolution of these two gaps has been studied[12] in detail for anisotropy up to $D/J=0.25$. Under the assumption that nothing significant happens in the presence of anisotropy, the excitation at $k=0$ and $S^z=\pm1$ should consist of one excitation with $k=\pi$, $S^z=0$ and one excitation with $k=\pi$, $S^z=\pm1$. Thus, the gap at $k=0$ and $S^z=\pm1$ should be equal to $G^{(+)}+G^{(-)}$.

To investigate the excitation spectrum we have used a Lanczös algorithm with minimized iterations applied to chains of up to 18 spins. An accuracy of 10^{-6} on the energy is obtained typically in a few tens of iterations. On a Cray2 computer, the lowest energy of the 18-spin chain sec obtained with such an accuracy in $\sim$3100 ($\sim$2400) secs for $S^z=0$ ($S^z=\pm1$) once the matrix elements are computed. In addition to the ground-state energy, we have computed the lowest energy of the sector $S^z=\pm1$ for all the allowed momenta. The difference ϵ_k is called the excitation energy ($\epsilon_\pi=G^{(-)}$). It has been obtained for all sizes from 4 to 16 sites (for 18 sites we have only computed the $k=0$ and π momenta around $D=0$ and J). The momentum $k=0$ and π are common to all these

chains. In these cases it is possible to use efficiently the so-called Shanks transformation[14] which is well suited to the removal of the exponential transients[4,12] that we expect in a massive chain (an estimate of the error can be obtained using a one-parameter family of transformations[15] which generalizes the Shanks transformation). In the isotropic $D=0$ case we find for the Haldane gap at $k=\pi$ the value 0.411(1) J. At $k=0$ we find a gap of 0.86(3) close to twice the Haldane value. Our findings for the isotropic chain are summarized in Fig. 1 (the finite-size effects are found to be more important for $k=0$ than for $k=\pi$). When adding anisotropy, we find that the gap at $k=0$ is given with a good accuracy by $G^{(+)}+G^{(-)}$. In the case of NENP, where $D/J\approx 0.2$, we have $G^{(+)}=0.684(1)$ while $G^{(-)}=0.289(1)$ giving a sum 0.973(2) which is very close to the measurement at $k=0$: 1.01(2) (in units of J). Up to $D/J\sim 1$, the sum property is well satisfied inside our errors.

The Haldane phase will survive until $G^{(-)}=0$. This closure of the lowest gap happens for $D=D_c\approx J$. In the lower part of Fig. 2, we have plotted the values of $G^{(-)}$ against the anisotropy term D using the Shanks transformation for 4–16 chains. The transition occurs clearly in a narrow region.[3,4,6] The most likely proposal[6] is that there is only one transition occurring in an isolated point. However, there may be a small massless phase with a nonzero width in D. The fact that the Shanks transformation can be used successfully near $D\sim J$ with 4–16 spin chains shows that the singular behavior shows up only for long chains. Our ability to use chains up to 18 sites can thus clarify the analysis.

We have thus computed the gap from $D/J=0.9$ up to 1.1 by steps of 0.01 for 4–16 chains and by steps of 0.02 for 18 chains (from $D/J=0.9$ up to 1.02). We have determined the transition point D_c and the critical indice ν by use of finite-size scaling with an unknown critical curve f:

$$NG_N^{(-)}=f[(D-D_c)N^{1/\nu}] . \tag{2}$$

The result is shown in Fig. 3, in the upper part for 14–18 chains and in the lower part for 10–18 chains. We find good evidence for scaling in the longer chains. The parameters are chosen to obtain visual matching of the points on a single curve. An analysis of the D dependence of logarithms of ratios of $NG_N^{(-)}$ for different N values[16] leads us to prefer a slightly reduced D_c central

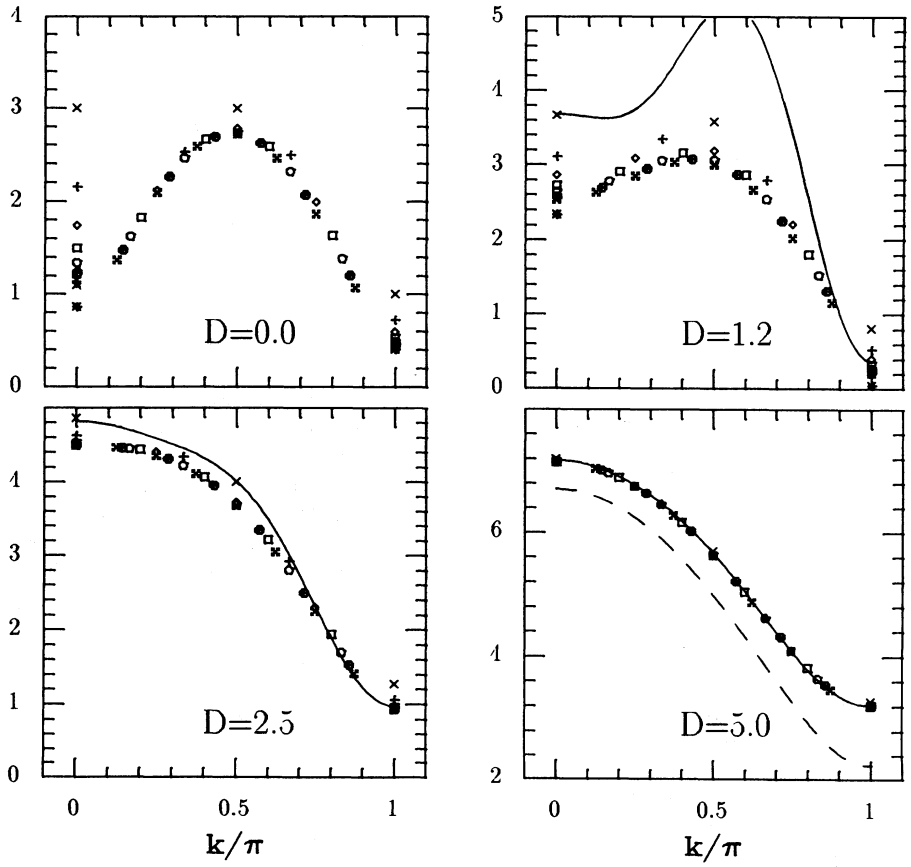

FIG. 1. Excitation spectrum as a function of the momentum for four values of the anisotropy. Data from $N=4$ (cross), 6 (plus), 8 (diamond), 10 (square), 12 (octagon), 14 (fancy plus), 16 (fancy cross), 18 (fancy square), and Shanks extrapolation (burst). The solid line is from Eq. (3) and the dashed line is from the semiclassical formula. D and ϵ are in units of J.

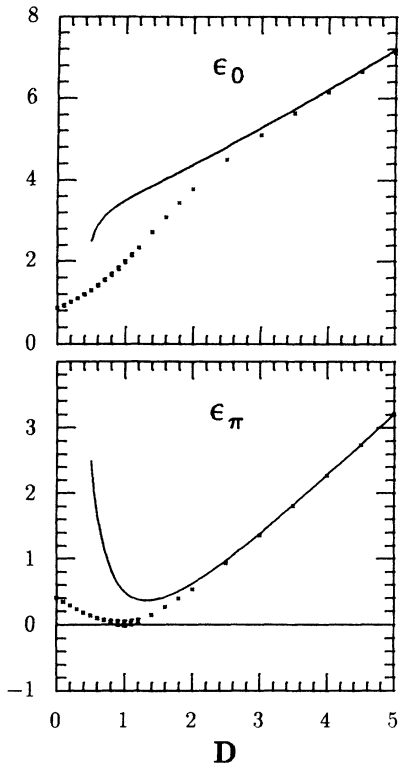

FIG. 2. Excitation ϵ_0 and ϵ_π as a function of the anisotropy, in units of J. The solid line is obtained from Eq. (3).

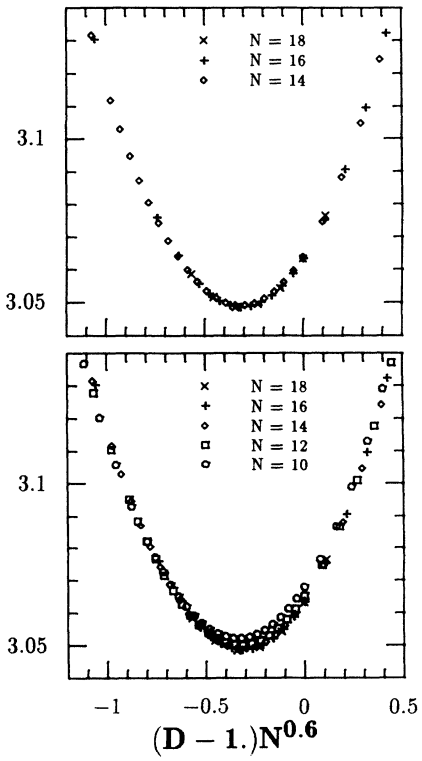

FIG. 3. Scaled gaps $NG_N^{(-)}$ as a function of the scaling variable in Eq. (2). Gaps and anisotropy are in units of J.

value. We conclude that the zero gap region reduced to an isolated point at $D_c/J=0.99(2)$ with a critical exponent $1/\nu=0.6(2)$. In the Abelian bosonization framework, the massless point is reached by setting to zero the coefficient of the relevant operator of a sine-Gordon model.[6] A simple scaling argument shows that the scaling dimension x of this operator is given by $x=2-(1/\nu)$. This, in turn, leads to a value for the exponent η governing the decay of correlations at the massless point: $\eta=1/2x\approx 0.36$ in excellent agreement with a direct determination.[6]

For larger D values, the gap rises again as seen in Fig. 2. For D not large enough the correlation length remains large and the finite-size effects are important on the spectrum as seen at Fig. 1 for $D/J=1.2$. The physics of this phase is quite different from the Haldane phase and can be understood from the large-D limit. When $D\to\infty$, the ground state becomes simply $|S_1^z=\cdots=S_N^z=0\rangle$ since all the spins are forced to lie in the XY plane. There is thus a gap of order D toward excitation of one spin to $S^z=\pm 1$. The states $|S_1^z=0,\ldots,S_i^z=\pm 1,\ldots,S_N^z=0\rangle$ will be dispersed under the action of the nearest-neighbor exchange term and form a band with $\epsilon_k=D+2J\cos k$ at first order in perturbation (in J). These states[7,9] with one up or down spin have been christened excitons (e) and antiexcitons ($\bar{e}$) since they are very different from magnon states in an antiferromagnet. They have been observed[9] in the compound $CsFeBr_3$ which has a very large single-ion anisotropy. Perturbation theory to third order has been performed[7] with the result

$$\epsilon_k = D\left[1+2\cos k\frac{J}{D}+(1+2\sin^2 k)\frac{J^2}{D^2} + [2\sin^2 k-\tfrac{1}{2}(1+8\sin^2 k)\cos k]\frac{J^3}{D^3}\right] + D\,O(J^4/D^4)\,. \tag{3}$$

We have investigated the dispersion relation for various values of the anisotropy in the large-D phase. As seen in Figs. 1 and 2, the agreement between our *ab initio* results and the perturbative result (solid line) is excellent for $D/J=5$ and 2.5. In these cases the gap is so large that we do not need any extrapolation method to estimate the infinite volume limit. It is only below $D\approx 2J$ that there are some significant deviations: for example, we display the spectrum ϵ_k for $D=1.2$ J, the immediate neighborhood of the transition which shows strong deviation from the perturbation theory. The gap value $G^{(-)}$ can be obtained from Eq. (3) and compared with numerical data: see Fig. 2 which summarizes our findings as a function of D. From our curves it is clear that the semiclassical formula derived by Lindgard,[17] $\epsilon_k=D\sqrt{1+4(J/D)\cos k}$, (dashed line in Fig. 1), is valid only asymptotically when $D\to\infty$ and that the straightforward perturbative result [Eq. (3)] is much more realistic for small-D values. This implies, that, in the case of $CsFeBr_3$, the anisotropy parameter should be[7] $D/J=3.3$ rather than[9] $D/J=4.7$ as suggested by use of the semiclassical result.

In conclusion, we have obtained evidence that, in the

whole Haldane phase of the anisotropic chain, the gap at $k=0$ is given by the sum of the two gaps at $k=\pi$. For the NENP compound we thus predict the $k=0$ $S^z=1$ gap to be ≈ 3.7 meV. At large anisotropy, perturbation theory is smoothly recovered as soon as $D/J>2.5$. The finite-size scaling analysis of the transition between the two phases leads to a critical value $D_c/J=0.99(2)$ with an exponent $1/\nu=0.6(2)$.

We thank A. V. Chubukov and A. W. W. Ludwig on the theoretical side and L. P. Regnault and J. Rossat-Mignod on the experimental side for discussions about the present status of neutron studies of NENP. We also thank M. Evans and T. Bhattacharya for a critical reading of our manuscript. The service de Physique Théorique is Laboratoire de la Direction des Sciences de la Matiére du Commissariat á l'Energie Atomique.

[1]F. D. M. Haldane, Phys. Rev. Lett. **50**, 1153 (1983); Phys. Lett. **93A**, 464 (1983).

[2]For a recent review see, I. Affleck, J. Phys.: Condens. Matter **1**, 3047 (1989).

[3]R. Botet, R. Jullien, and M. Kolb, Phys. Rev. B **28**, 3914 (1983).

[4]T. Sakai and M. Takahashi, Phys. Rev. B **42**, 1090 (1990); **42**, 4537 (1990).

[5]M. Takahashi, Phys. Rev. Lett. **62**, 2313 (1989).

[6]H. J. Schulz and T. Ziman, Phys. Rev. B **33**, 6545 (1986); H. J. Schulz, *ibid.* **34**, 6372 (1986).

[7]N. Papanicolaou and P. Spathis, J. Phys.: Condens. Matter **1**, 5555 (1989); **2**, 6575 (1990).

[8]L. P. Regnault, J. Rossat-Mignod, and J. P. Renard (unpublished).

[9]B. Dorner *et al.*, Z. Phys. B **72**, 487 (1988).

[10]J. P. Renard *et al.*, Europhys. Lett. **3**, 945 (1987); K. Katsumata *et al.*, Phys. Rev. Lett. **63**, 86 (1989).

[11]T. Delica, K. Kopinga, H. Leschke, and K. K. Mon, Europhys. Lett. **15**, 55 (1991).

[12]O. Golinelli, Th. Jolicoeur, and R. Lacaze, Phys. Rev. B **45**, 9798 (1992).

[13]I. Affleck and R. A. Weston, Phys. Rev. B **45**, 4667 (1992).

[14]D. Shanks, J. Math. Phys. (N.Y.) **34**, 1 (1955).

[15]J. M. Van den Broeck and L. W. Schwartz, SIAM J. Math. Anal. **10**, 658 (1979).

[16]U. Glaus and T. Schneider, Phys. Rev. B **30**, 215 (1984).

[17]P. A. Lindgard, Physica B **120**, 190 (1983).