Large-Scale Numerical Evidence for Bose Condensation in the $S = 1$ Antiferromagnetic Chain in a Strong Field

Erik S. Sørensen[1] and Ian Affleck[1,2]
[1]*Department of Physics, University of British Columbia, Vancouver, British Columbia, Canada V6T 1Z1*
[2]*Canadian Institute for Advanced Research, University of British Columbia, Vancouver, British Columbia, Canada V6T 1Z1*
(Received 16 June 1993)

Using the recently proposed density matrix renormalization group technique we show that the magnons in the $S = 1$ antiferromagnetic Heisenberg chain effectively behave as bosons that condense at a critical field h_c. We determine the spin-wave velocity, $v = 2.49(1)$, as well as the gap $\Delta = 0.4107(1)J$.

PACS numbers: 75.10.Jm, 75.40.Mg

It is by now well established both experimentally [1] and theoretically [2–7] that the $S = 1$ antiferromagnetic chain has a gap, Δ, to a triplet excitation above a singlet ground state. Thus the magnetization M remains strictly zero up to a critical field $h_c = \Delta$. For the quasi-one-dimensional system NENP the critical field is found to be about 10 T [8] for a field applied along the symmetry axis. The behavior of $M(h)$ just above h_c has been the subject of some theoretical work [9–11]. The existing experimental results [12] appear to be dominated by extraneous effects such as an off-diagonal alternating component of the gyromagnetic tensor [13], interchain coupling, anisotropies, and impurities.

This problem can be solved using a boson quasiparticle model involving triplet magnons with repulsive interactions for parallel spins [11]. This model predicts that $h_c = \Delta$. (We set the Bohr magneton and g factors to 1.) At this field the "rest-mass energy" of the magnons is exactly canceled by their Zeeman energy and one-dimensional Bose condensation occurs. What prevents a catastrophe from occurring at h_c is the intermagnon repulsion which governs the behavior of $M(h)$ above h_c. To calculate $M(h)$ we need to calculate the energy $E(M)$ of a very dilute system of M polarized magnons in their ground state. $M(h)$ is then found by inverting the equation $h = dE/dM$. It was argued in Ref. [11] that the behavior of $E(M)$ is the same as for a system of nonrelativistic noninteracting *fermions*:

$$E = (\Delta - h)M + L\int_{-k_F}^{k_F} \frac{dk}{2\pi}\frac{v^2k^2}{2\Delta}. \tag{1}$$

Here v is the velocity, determined from the single magnon dispersion relation at low energies:

$$\omega(k) = \Delta + v^2(k-\pi)^2/2\Delta + O([k-\pi]^3), \tag{2}$$

L is the length of the system, and k_F is determined from the particle number:

$$M = L\int_{-k_F}^{k_F}\frac{dk}{2\pi} = Lk_F/\pi. \tag{3}$$

This gives $E = M(\Delta - h) + (v\pi)^2M^3/6\Delta L^2 + O(M^4)$ and hence

$$M/L = \sqrt{(h-\Delta)2\Delta}/\pi v, \tag{4}$$

up to terms of higher order in $h - \Delta$.

This formula was first given based on a noninteracting fermion model of magnons [9]. It was later argued to also arise from an *interacting boson* model [11] sufficiently close to h_c, and hence to be exact. It is expected to be valid for very general short-range repulsive interactions between the spin-polarized magnons. It should hold for arbitrarily weak interactions for long enough chains. The reason is that when the average intermagnon spacing is very large compared to the range of the repulsive interaction the multimagnon wave function, $\Psi_M(x_1, x_2, ..., x_M)$, can be approximated by a free fermion (Bloch) wave function multiplied by the sign function $\epsilon(x_1, x_2, ...x_M)$ which has the value ± 1 and changes sign whenever two particles are interchanged:

$$\Psi_M(x_1, x_2, ..., x_M) = \frac{1}{\sqrt{M!}}\epsilon(x_1, x_2, ..., x_M)\sum_{P(i_1\cdots i_M)}\prod_{i=1}^{M}\psi_1(x_{i_1})\psi_2(x_{i_2})\cdots\psi_M(x_{i_M})\mathrm{sgn}P. \tag{5}$$

Here P denotes the permutation and sgn$P$ the sign of the permutation. $\psi_i(x)$ is a single-particle noninteracting wave function depending on the wave vector k_i. Ψ_M is symmetric as required by Bose statistics, is a solution of the noninteracting Schrödinger equation almost everywhere (i.e., except where two or more particles intersect), and vanishes whenever two or more particles come together. As such it is expected to become exact in the dilute limit and hence to give exactly the magnetization as $h \to h_c$.

To simulate the M-magnon problem in the dilute regime we need a length $L \gg M\xi$ where $\xi \approx 6$ is the

correlation length or the approximate range of the intermagnon interaction. In practice it appears that $L \geq 30M$ is required. Before last year, the longest chains that had been studied accurately had $L = 32$, so good results were only available for a single magnon and it was impossible to study magnon interactions. Thanks to a breakthrough in the real-space renormalization group technique made by White [2] it is now feasible to study chains of length 100 or longer using a density matrix renormalization group (DMRG) approach. We will present results here on chains of length up to 100 containing up to three magnons. White and Huse [5] obtained related numerical results independently. We analyze our results in a different way which establishes Eq. (4). Our results indicate quite convincingly that the lowest-energy two or three magnon state has the form discussed above; namely, the lowest-energy free fermion wave function multiplied by the sign function. We establish this result in two ways. First, we study $S^z(x) = \sum_{j=1}^{M} \delta(x - \hat{x}_j)$, where $\hat{x}_j$ is the position operator for the jth particle, showing that it has the expected form

$$\langle S^z(x) \rangle = M \int dx_2 dx_3 \cdots dx_M |\Psi_M(x, x_2, x_3, ..., x_M)|^2. \tag{6}$$

Second, we study the finite-size dependence of the energy of the multimagnon ground state, showing that it behaves as

$$E(M) \approx \sum_{i=1}^{M} \omega(k_i). \tag{7}$$

Holding M fixed and taking $L \to \infty$, the k_i's are $O(1/L)$ so the above formula gives $E(M, L) = M(\Delta - h) + a(M)/L^2$. The corrections to Eq. (7) from intermagnon interactions are expected to be $O(L^{-3})$. This power of L can be most easily understood in the $M = 2$ case. The wave function contains a L^{-1} normalization factor but is $O(L^{-2})$ over the entire region where the interaction is non-negligible since it vanishes proportional to $k(x_1 - x_2)$ with k of $O(L^{-1})$. The power L^{-3} arises from squaring the wave function and picking up a factor of L from the integral over the center-of-mass position. We verify that the corrections to Eq. (7) are indeed of this order.

Implementing the DMRG optimally requires studying chains with open, rather than periodic, boundary conditions. Such chains have $S = 1/2$ excitations localized near the ends which have been the subject of a number of experimental and theoretical studies [14]. For our purposes they are just a minor annoyance. They are also found to have repulsive interactions with the bulk magnons for parallel spins. Thus the magnon wave functions essentially obey vanishing boundary conditions at the ends, with additional corrections of $O(L^{-3})$ to the energy, Eq. (7), from the magnon-end excitation interactions. We also considered more general boundary conditions for the magnon wave functions. These were found to change the energy only to $O(L^{-3})$ since they lead to negligible changes in the wave function.

The DMRG method for open chains leaves only two good quantum numbers: the total S^z component, S^z_T, and the parity, P. We need to determine the parity for low-lying states with a given S^z_T. We shall only be concerned with chains of even length. For these chains the ground state is a singlet with even parity, 0^+. Above the ground state is an exponentially low-lying triplet, 1^-. In the thermodynamic limit the triplet and the singlet become degenerate and the ground state fourfold degenerate. This spectrum can be seen to arise from the two $S = 1/2$ end excitations forming either an odd parity singlet or an even parity triplet, in addition to an overall parity flip coming from the rest of the ground state. This parity flip can be understood from the valence bond solid state [7] where we draw two valence bonds emanating from each site. These valence bonds represent singlet contractions of pairs of $S = 1/2$ spins so they have a directionality associated with them. When we make a parity transformation we flip the orientation of an odd number of valence bonds resulting in a $(-)$ sign. Thus, the parity, P_E, of a state with no magnons present is $(+)$ if the end excitations combine into the singlet and $(-)$ for the triplet. The parity of higher excited states, containing one or more magnons, is a product of three factors, $P_E P_{SW} P_m$. P_m contains a contribution of $(-)$ from each magnon present. This is because the magnons are created and annihilated by the staggered magnetization operator, and this changes sign upon switching even and odd sublattices. P_{SW} is the parity of the spatial wave functions of the magnons. For instance, for a single magnon, the wave functions, ψ_i, in Eq. (5) are $\psi_i = \sqrt{2/(L-1)} \sin k_i x$, $k_i = \pi n_i/(L-1)$, with n_i *odd* for even parity and n_i *even* for odd parity. We take $0 \leq x \leq L - 1$ and parity will therefore take x into $L - 1 - x$.

For a chain with open boundary conditions the lowest lying state of a given magnetization, M, will have $M = m + 1$, where m is the number of magnons present and the additional term, 1, corresponds to the end excitations forming a triplet. In order to minimize the intermagnon repulsion, the wave function for large L takes the Bloch form of Eq. (5), with ψ_i as above and $n_i = i$, in order to satisfy vanishing boundary conditions. Thus Eq. (1) becomes

$$E_{m+1}(L) - E_1(L) = (\Delta - h)m + \frac{(v\pi)^2}{2\Delta(L-1)^2} \sum_{i=1}^{m} n_i^2 + O(L^{-3}), \quad n_i = i. \tag{8}$$

We test this formula below for $m = 1$, 2, and 3.

The 1^- state becomes degenerate with the ground state in the thermodynamic limit, exponentially fast with L, and we shall therefore use it as the reference state and calculate energy gaps with respect to this state and not 0^+, as already implied in Eq. (8). We have calculated the gap as a function of chain length between this state and three of the low-lying states using density matrices of the

TABLE I. The spectrum of the $L = 100$ open $S = 1$ antiferromagnetic Heisenberg chain.

S_T^P	$-E$
1^-	138.940 086
2^+	138.522 461
3^-	138.085 57
4^+	137.603

size 243 × 243 keeping 81 eigenvectors of these matrices at each iterations. For each of these states we have also calculated $\langle S_i^z \rangle$ and $\langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle$ along a $L = 100$ site chain using a finite lattice method [2]. A brief summary of our results is shown in Table I.

The lowest-lying $M = m + 1 = 2$ state corresponds to a state with the end excitations in the 1^- state and one magnon present. This state has therefore parity (+), since $P_E = (-), P_{SW} = (+)$, and $P_m = (-)$. We approximate the wave function as consisting of two factors existing in different Hilbert spaces: a factor from the end excitations and a factor, Ψ_m, corresponding to the m-magnon wave function. From the above discussion we see that for a single magnon the magnon part of the wave function, Ψ_m, becomes $\Psi_1 = \sqrt{2/(L-1)} \sin k_1 x, \quad n_1 = 1$. From Eq. (6) we then see that $\langle S_i^z \rangle_{2^+} - \langle S_i^z \rangle_{1^-} = \frac{2}{L-1} \sin^2 k_1 x$, which is shown as the solid line in Fig. 1. Here the subtraction of $\langle S_i^z \rangle_{1^-}$ essentially removes any contribution from the end excitations. An excellent agreement is evident. Also shown in Fig. 1 is the local bond energy $e_i^{21} = \langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle_{2^+} - \langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle_{1^-}$. The dilute boson model predicts an energy density, $e(x) \simeq \Delta \sum_{j=1}^{m} \delta(x - \hat{x}_j)$ ignoring the $O(L^{-2})$ kinetic energy. Therefore $e(x)$ should be proportional to $S^z(x)$ in Eq. (7), the proportionality factor being the gap, Δ. In Fig. 1 this prediction is shown as the dashed line. From Eq. (8) we can now extract values for the gap, Δ, and the velocity, v. The fit of $\Delta_{21}(L) = E_{2^+}(L) - E_{1^-}(L)$ by Eq. (8) is excellent, and we obtain $\Delta_{21}(L) = 0.4107(1) + 74.7(4)(L-1)^{-2} + O([L-1]^{-3})$, with $\chi^2 = 4.55$. We see that $\Delta = 0.4107(1)$, $v = 2.49(1)$. The value of Δ is in excellent agreement with what was previously obtained [2,5]. The value of v is in good agreement with the value $v = 2.46$ that can be extracted from exact diagonalization [6], and the value $v \sim 2.36$ obtained from $1/S$ expansions [15]. It is also in good agreement with the experimental results on NENP [16], $v \sim 2.45$. The coefficient, 74.7(4), in front of the $(L-1)^{-2}$ term, which determines v, differs marginally from what was obtained by White [2], (67.9), due to the use of a different polynomial form.

The lowest-lying 2-magnon state has parity (−) since $P_E = (-)$, $P_{SW} = (+)$, $P_m = (+)$, and total magnetization $M = m + 1 = 3$. The magnon part of the wave function is $\Psi_2 = \frac{2}{L-1}[\sin k_1 x_1 \sin k_2 x_2 - \sin k_1 x_2 \sin k_2 x_1]\epsilon(x_1, x_2)$ with $n_1 = 1$, $n_2 = 2$. Note that under parity $x_i \to L - x_i - 1$, $\sin k_1 x_i$ is even, $\sin k_2 x_i$ is odd, and $\epsilon(x_1, x_2)$ is odd, resulting in $P_{SW} = (+)$. We now obtain $\langle S_i^z \rangle_{3^-} - \langle S_i^z \rangle_{1^-} = \frac{2}{L-1}\{\sin^2 k_1 x + \sin^2 k_2 x\}$, which is shown as the solid line in Fig. 2. The dashed line represents the theoretical prediction for the local bond energy which is also shown in Fig. 2. Again we fit $\Delta_{31}(L) = E_{3^-}(L) - E_{1^-}(L)$ by Eq. (8) and we obtain $\Delta_{31}(L) = 0.823(1) + 359(5)(L-1)^{-2} + O([L-1]^{-3})$. The constant term should be 2Δ, in good agreement with the value of Δ obtained above. Since in this case we expect $n_1 = 1, n_2 = 2$, and therefore $\sum n_i^2 = 5$, the coefficient in front of the $(L-1)^{-2}$ term should be 5 times greater than what we found for the 2^- level. Clearly this is the

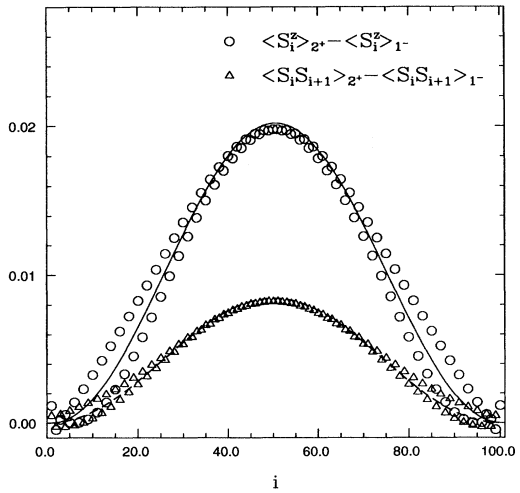

FIG. 1. The open circles represent $\langle S_i^z \rangle_{2^+} - \langle S_i^z \rangle_{1^-}$. The solid line is the expression given in the text. Also shown, by triangles, is $\langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle_{2^+} - \langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle_{1^-}$. The dashed line is the prediction for this local bond energy.

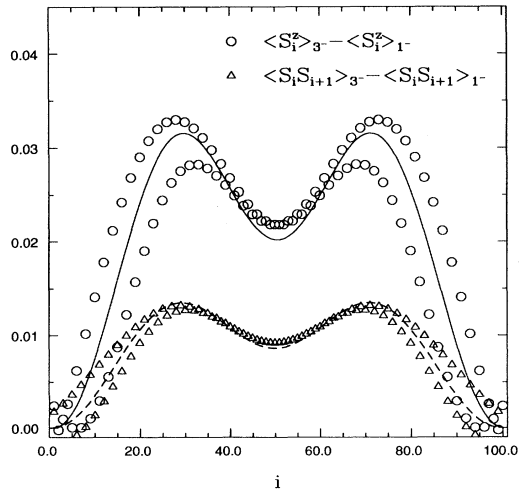

FIG. 2. The open circles represent $\langle S_i^z \rangle_{3^-} - \langle S_i^z \rangle_{1^-}$. The solid line is the expression given in the text. Also shown, by triangles, is $\langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle_{3^-} - \langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle_{1^-}$. The dashed line is the prediction for this local bond energy.

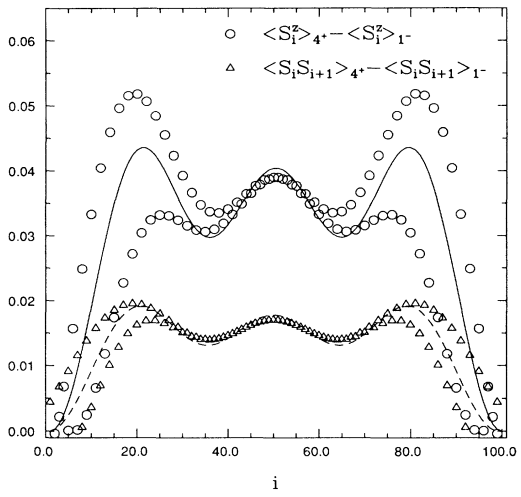

FIG. 3. The open circles represent $\langle S_i^z\rangle_{4+} - \langle S_i^z\rangle_{1-}$. The solid line is the expression given in the text. Also shown, by triangles, is $\langle \mathbf{S}_i \cdot \mathbf{S}_{i+1}\rangle_{4+} - \langle \mathbf{S}_i \cdot \mathbf{S}_{i+1}\rangle_{1-}$. The dashed line is the prediction for this local bond energy.

case: We obtain $\sum n_i^2 = 4.80(6)$, and the only values of n_i consistent with our results are indeed $n_1 = 1, n_2 = 2$.

The three magnon state with $M = m + 1 = 4$ has parity (+) by the same arguments as above. The wave function now has six terms and we obtain $\langle S_i^z\rangle_{4+} - \langle S_i^z\rangle_{1-} = \frac{2}{L-1}\{\sin^2 k_1 x + \sin^2 k_2 x + \sin^2 k_3 x\}$, with $n_1 = 1, n_2 = 2, n_3 = 3$. This expression is shown as the solid line in Fig. 3, along with the numerical results for $\langle S_i^z\rangle_{4+} - \langle S_i^z\rangle_{1-}$ and the local bond energy $\langle \mathbf{S}_i \cdot \mathbf{S}_{i+1}\rangle_{4+} - \langle \mathbf{S}_i \cdot \mathbf{S}_{i+1}\rangle_{1-}$. The dashed line is the prediction for the local bond energy. Again good agreement is evident between theory and the numerical results. Fitting the energy gap to Eq. (8) we find that the $(L-1)^{-2}$ term now has a coefficient of 1030(150). Thus in this case, if we use the value of v determined above, $\sum n_i^2 = 14(2)$. This is only consistent with the values $n_1 = 1, n_2 = 2, n_3 = 3$. We also performed some calculations for $L = 60$; these show that effects from the end excitations clearly diminish as L is increased.

In summary, we find that the numerical results are in excellent agreement with Eq. (8) with $v = 2.49(1)$, $\Delta = 0.4107(1)$. The magnons behave as bosons with repulsive interactions among themselves and with the end excitations. The lowest energy state for given M has the Bloch form of Eq. (5), implying the validity of Eq. (4).

We would like to thank Steven White for sending us Refs. [2] and [5] prior to publication as well as for useful discussions. This research was supported in part by NSERC of Canada.

[1] W. J. L. Buyers, R. M. Morra, R. L. Armstrong, M. J. Hogan, P. Gerlach, and K. Hirikawa, Phys. Rev. Lett. **56**, 371 (1986); J. P. Renard, M. Verdaguer, L.P. Regnault, W. A. C. Erkelens, J. Rossat-Mignod, and W. G. Stirling, Europhys. Lett. **3**, 945 (1988).
[2] S. R. White, Phys. Rev. Lett. **69**, 2863 (1992).
[3] F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983); R. Botet and R. Julien, Phys. Rev. B **27**, 613 (1983); R. Botet, R. Julien, and M. Kolb, Phys. Rev. B **28**, 3914 (1983); M. Kolb, R. Botet, and R. Julien, J. Phys. A **16** L673 (1983); M. P. Nightingale and H. W. J. Blöte, Phys. Rev. B **33**, 659 (1986); H. J. Schulz and T. A. L. Ziman, Phys. Rev. B **33**, 6545 (1986).
[4] M. Takahashi, Phys. Rev. Lett. **62**, 2313 (1989).
[5] S. R. White and D. A. Huse (to be published).
[6] M. Takahashi (to be published).
[7] For a review see I. Affleck, J. Phys. Condens. Matter **1**, 3047 (1989).
[8] Y. Ajiro, T. Goto, H. Kikuchi, T. Sakakibara, and T. Inami, Phys. Rev. Lett. **63**, 1424 (1989); K. Katsumata, H. Hori, T. Takeuchi, M. Date, A. Yamagishi, and J. P. Renard, Phys. Rev. Lett. **63**, 86 (1989).
[9] A. M. Tsvelik, Phys. Rev. B **42**, 10 499 (1990).
[10] K. Nomura and T. Sakai, Phys. Rev. B **44**, 5092 (1991); T. Sakai and M. Takahashi, Phys. Rev. B **43**, 13 383 (1991).
[11] I. Affleck, Phys. Rev. B **43**, 3215 (1991).
[12] K. Katsumata, H. Hori, T. Takeuchi, M. Date, A. Yamagishi, and J.P. Renard, Phys. Rev. Lett. **63**, 86 (1989); Y. Ajiro, T. Goto, H. Kikuchi, T. Sakakibara, and T. Inami, Phys. Rev. Lett. **63**, 1424 (1989).
[13] M. Chiba, Y. Ajiro, H. Kikuchi, T. Kubo, and T. Morimoto, Phys. Rev. B **44**, 2838 (1991).
[14] T. Kennedy, J. Phys. Condens. Matter **2**, 5737 (1990); M. Hagiwara, K. Katsumata, I. Affleck, B. I. Halperin, and J.-P. Renard, Phys. Rev. Lett. **65**, 3181 (1990); S. J. Glarum, S. Geschwind, K. M. Lee, M. L. Kaplan, and J. Michel, Phys. Rev. Lett. **67**, 1614 (1991).
[15] D. V. Kveshchenko and A. V. Chubukov, Zh. Eksp. Teor. Fiz. **93**, 1904 (1987) [Sov. Phys. JETP **66**, 1088 (1987)].
[16] S. Ma, C. Broholm, D. H. Reich, B. J. Sternlieb, and R. W. Erwin, Phys. Rev. Lett. **69**, 3571 (1992).