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Theory of Haldane-gap antiferromagnets in applied fields

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As first predicted by Haldane, and later observed in neutron-scattering experiments, one-dimensional Heisenberg antiferromagnets of integer spin have an excitation gap. Recently, the highly one-dimensional antiferromagnet $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ has been studied in an applied magnetic field. We discuss the susceptibility, high-field magnetization, and field-dependent neutron scattering using general arguments and field-theory methods.

I. INTRODUCTION

Haldane first argued[1] that spin-s one-dimensional Heisenberg antiferromagnets have an excitation gap and a finite correlation length for an integer (but not a half-integer) s. This is by now quite well established theoretically based on finite-size calculations, field-theory arguments, and an exactly solvable model.[2] The first experimental evidence for the Haldane gap came from neutron-scattering experiments[3] on the $s=1$ system $CsNiCl_3$. While this material is highly isotropic in its spin couplings, it is only moderately one dimensional. (The ratio of interchain to intrachain couplings is estimated to be 0.017.) Consequently, there is Néel order at low temperatures and significant planar dispersion in the excitation spectrum. $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ (NENP) (also of spin 1) is much more one dimensional (the ratio of couplings is estimated to be 0.0006), and it appears to be disordered even at $T=0$. However, it has quite significant planar anisotropy. The lowest excited state, predicted by Haldane to be a triplet of total spin 1 (above a spin-zero ground state) is split into a doublet and a singlet with the ratio of excitation energies observed in neutron-scattering experiments[4] to be about 1:2. Apart from zero-field neutron-scattering, a number of other measurements have recently been made in nonzero applied magnetic fields. These include susceptibility,[4] high-field magnetization,[5-7] and neutron scattering in a finite field.[5] In all these measurements the anisotropy is quite evident, as is the Haldane gap.

The field-theory treatment of the system is based on the Lorenz invariant O(3) nonlinear σ model, which is equivalent to the low-energy limit of the Heisenberg model at large s. This is a highly nontrivial field theory whose spectrum consists of a triplet of bosons with nonzero scattering but no bound states. Although the exact S matrix is known for this model, nothing directly amenable to experiment is exactly calculable. The O(n) model is easily solvable in the large-n approximation, which seems to be at least qualitatively reliable for $n=3$. In this approximation, the model essentially reduces to a triplet of free massive boson fields, $\boldsymbol{\varphi}$. This provides a type of exactly solvable mean-field theory for Haldane gap antiferromagnets, which seems to contain most of the essential physics. The mean-field theory was applied recently[8] to studying the effects of interchain couplings. A φ^4 coupling was added for stability, in the spirit of a Landau-Ginsburg model. Since the σ model is only exact for large s, it is not clear which model is better for $s=1$.

In this paper we wish to develop a theoretical picture of Haldane gap antiferromagnets in an applied magnetic field. The results are divided into two sections. Those in Sec. II follow from general principles, well-controlled numerical calculations, or order of magnitude estimates. The results in Sec. III depend on the mean-field theory. Section IV contains conclusions and suggestions for more work, both experimental and calculational.

II. GENERAL RESULTS

NENP is believed to be well represented by the spin-1 Heisenberg model with easy-plane crystal-field anisotropy:

$$H=J\sum_i \mathbf{S}_i\cdot\mathbf{S}_{i+1}+D\sum_i (S_i^z)^2,\quad J\approx 48\mathrm{K}, D\approx 12\ \mathrm{K}\ . \tag{2.1}$$

Numerical simulations for $D=0$ indicate a correlation length of about five lattice spacings, a gap of about $\Delta=0.4J$, and a spin-wave velocity of about $v=2.56J$ (28% higher than the lowest-order spin-wave theory result) (Refs. 2 and 9). The ground state has a total spin $\mathbf{S}=0$, and the lowest excitation is a triplet ($s=1$) with dispersion relation near wave vector π of

$$E\approx[v^2(k-\pi)^2+\Delta^2]^{1/2}\ . \tag{2.2}$$

Near $k=0$ the gap is observed[9] to be about 2Δ. For finite positive D the triplet splits into a higher-energy singlet and a lower-energy doublet with the quantum numbers of

$$|a\rangle\equiv\sum_x e^{ikx}S_x^a|0\rangle\ , \tag{2.3}$$

where $a=z$ for the singlet, $z=x$ or y for the doublet, and $|0\rangle$ is the singlet ground state. Note that $|z\rangle$ has total z component of spin, $S^z=0$, whereas the states,

$$|\pm\rangle \equiv (|x\rangle \pm i|y\rangle)/\sqrt{2}\ , \tag{2.4}$$

have $S^z = \pm 1$, respectively. The gaps observed in neutron scattering[4] are approximately $\Delta z \approx 30$ K and $\Delta_\pm \approx 14$ K. (In NENP the z axis is the b axis in crystalographic notation.) An applied magnetic field, h, in the i direction leads to the additional term in the Hamiltonian:

$$H \to H - g_i \mu_B h S^i\ . \tag{2.5}$$

Here the g factors are believed to be $g_z \approx 2.15$ and $g_{x,y} \approx 2.22$. The effect of a field in the z direction can be completely understood by symmetry arguments alone, since S^z is a conserved operator. A field in the x direction has effects that are more model dependent, since S^x is not conserved when the D term is present.

Let us begin with a field in the z direction. All states can be classified by their S^z quantum number, and their energies are shifted by

$$E \to E - g_z \mu_B h S^z\ . \tag{2.6}$$

(We emphasize again, this is an *exact* result.) Thus the higher $|z\rangle$ state is unaffected, whereas the doublet $|\pm\rangle$ splits by $\pm g_z \mu_B h$. For large enough h, one or more of the excited states will cross the ground state. In principle, the state that first crosses need not be the state $|+\rangle$; a higher-energy state of greater S^z might cross first. However, the field-theory treatment (using either the linear or nonlinear model) predicts that the complete spectrum consists only of multiparticle scattering states of the triplet. This implies that the lowest-energy state of given $S^z = n$, is simply a state of n well separated $|+\rangle$ excitations. It seems very likely that the lowest-energy states of given S^z in the spin chain are of this type. This is supported by the fact that the gap at $k=0$ is measured to be 2Δ, corresponding to a two-particle state, both particles carrying $k=\pi$. Thus when h is increased beyond a critical field of

$$h_c = \Delta_\pm / g_z \mu_B\ , \tag{2.7}$$

$|+\rangle$ excitations bose condense in the ground state. The gap, $\Delta_\pm$ obtained by this formula from the experimentally observed [7]h_c of 9.8 T is 14.2 K, in excellent agreement with the value of 14 K obtained directly from zero-field neutron scattering.[4] [A smaller h_c of about 8 T was reported in Ref. 6.] This provides strong support for the preceding description of the spectrum. Note that it is not necessary to postulate some independent dispersion relation near $k=0$; the gap there is simply $2(\Delta_\pm - g_z\mu_B h)$. Also note that it is $\Delta_\pm$ that determines h_c, not Δ_z, a point that may be slightly confusing, since the field is applied in the z direction.

The free boson model becomes clearly inadequate for $h > h_c$, since then the density of bosons in the ground state would be infinite. However, the repulsion between the bosons in the interacting theory leads to a finite density. We may express the energy of the ground state with a density, n, of + magnons as some function $E(n)$. It should have a Taylor expansion of the form

$$E(n)/L = (\Delta_\pm - g_z \mu_B h) n + \alpha n^2 + \beta n^3 \cdots\ , \tag{2.8}$$

where α is a positive constant and L is the number of spins on the chain. The magnetization per spin, $M = g_z \mu_B n$, is determined by minimizing $E(n)$. Thus this is strictly zero (at $T=0$) for $h < h_c$ and has a slope determined by α for h slightly bigger than h_c:

$$M \approx g_z \mu_B (g_z \mu_B h - \Delta_\pm)/2\alpha\ . \tag{2.9}$$

Note that this Bose condensation transition will get smoothed out at any finite T by standard thermodynamic arguments for one-dimensional systems.

The zero-field susceptibility per spin is given by

$$\chi^z = (g_z \mu_B)^2 \langle (S^z)^2 \rangle / LT\ , \tag{2.10}$$

where

$$\langle (S^z)^2 \rangle = (1/Z) \sum_i (S^z)^2 e^{-E_i/T}\ , \tag{2.11}$$

the sum is over all (multiparticle) states, and Z is the partition function. (We set Boltzmann's constant to one.) The lowest states of nonzero S^z are the single-particle states $|\pm\rangle$, so at $T \to 0$,

$$\chi^z \to e^{-\Delta_\pm/T}\ . \tag{2.12}$$

In fact we can make a more precise asymptotic prediction using the asymptotic form for the dispersion relation for the doublet:

$$E = \Delta_\pm^2 + v^2 (k-\pi)^2 / 2\Delta_\pm^2 + O((k-\pi)^4)\ . \tag{2.13}$$

[This formula should be exact, whereas the Lorentz invariant form of Eq. (2.2) for larger $(k-\pi)$ is only approximate. Equation (2.13) is essentially just a definition of v. v can be expected to depend somewhat on anisotropy and to be different for $\pm$ and z modes.] We thus predict

$$\chi^z \to (g_z \mu_B)^2 e^{-\Delta_\pm/T} 2 \int_{-\infty}^{\infty} (dk/2\pi) \exp(-v^2 k^2 / 2\Delta_\pm^2 T) = (g_z \mu_B)^2 e^{-\Delta_\pm/T} 2(\Delta_\pm / v)\sqrt{2\pi T}[1 + O(T/\Delta_\pm)]\ . \tag{2.14}$$

Note, in particular, that χ^z goes to zero at $T \to 0$. A nonzero $\chi^z(0)$ in a Haldane gap system indicates a breaking of the symmetry of rotation about the z axis, i.e., a splitting of the doublet $|\pm\rangle$. Indeed, there seems to be some contradiction between the absence of any observed splitting of the doublet in the neutron-scattering experiment and the nonzero measured value of χ^z. This problem is especially severe because it follows on very general grounds that

$$\chi^z(0) \propto (\Delta_+ - \Delta_-)^2\ . \tag{2.15}$$

When this rotational symmetry is broken, the exact formula for $\chi^z(0)$ becomes

$$\chi^z(0)=(g_z\mu_B)^2\sum_{i'}|\langle 0|S^z|i\rangle|^2/(E_i-E_0)\ , \tag{2.16}$$

where the sum is over all states other than the ground state, $|0\rangle$. In the symmetric case $S^z|0\rangle=0$, so $\chi^z(0)=0$. Consider some perturbation, H', which breaks the rotation symmetry [perhaps some crystal-field term like $D_x\sum_i(S_i^x)^2$]. In general, such a perturbation will produce a first-order splitting of the doublet,

$$\langle i|H'|i\rangle\neq 0\ , \tag{2.17}$$

for $|i\rangle,|j\rangle$ elements of the doublet. It also produces a second-order effect in $\chi^z(0)$:

$$\chi^z(0)\approx(g_z\mu_B)^2\sum_{i'}|\langle 0|H'|i\rangle|^2(S_i^z)^2/(E_i-E_0)^3\ . \tag{2.18}$$

The sum gets contributions from the single-particle states $|\pm\rangle$ as well as multi-particle states. Thus we could estimate the ratio of $T=0$ susceptibilities:

$$\chi^z(0)/\chi^x(0)\approx(\Delta_x-\Delta_y)^2/(\Delta_z-\Delta_\pm)^2\ . \tag{2.19}$$

The splitting of Δ_x and Δ_y based on the width[4] of the lower neutron scattering peak, $|\Delta_x-\Delta_y|\langle\Delta_\pm/10$, would imply $\chi^z(0)/\chi^x(0)<0.01$, whereas the measurements in Ref. 4 give a ratio of about 1/3. One possible explanation of this is that there is a third peak that has not, so far, been observed in the neutron-scattering cross section, and the breaking of the planar symmetry is substantial. Alternatively, perhaps the reported value of $\chi^z(0)$ is caused by extraneous effects. Indeed a second measurement of $\chi^z(0)$, in Ref. 7 seems to give a much smaller value. The discrepancy could be caused by difficulties in subtracting off the diamagnetic contribution or by impurities.

We now consider the effect of a field in the x direction (assuming again symmetry of rotation about the z axis only). The $T=0$ susceptibility is now nonzero and given by Eq. (2.16) with $z\to x$. The exact finite-T formula is

$$\chi^x(T)=(1/Z)(g_x\mu_B)^2\sum_{i,j}|\langle i|S^x|j\rangle|^2(e^{-E_i/T}-e^{-E_j/T})/(E_i-E_j)\ . \tag{2.20}$$

At low temperatures this has the form

$$\chi^x(T)=\text{const}+O(E^{-\Delta_\pm/T})\ . \tag{2.21}$$

Note that it is the lowest gap $\Delta_\pm$, not Δ_z, that appears here, since S^x has nonzero matrix elements involving the states $|\pm\rangle$, e.g., $\langle 0|S^x|\pm\rangle$. We know of essentially no exact results concerning finite fields in the x direction. Again the doublet should split, and the energy of the singlet, Δ_z should also change; none of these energies is expected to depend linearly on h. It seems unlikely that there will be a real phase transition even at $T=0$ in this case. In the free boson model we find that the energy of $|x\rangle$ remains fixed, whereas the energy of $|y\rangle$ decreases and Δ_z increases. Furthermore, a phase transition *does* occur at a critical h when the energy of $|y\rangle$ reaches 0. However, we suspect that the effects of interactions would lead to some shift of Δ_x and smooth out the phase transition. The observed field dependence of the neutron scattering[5] and the sharp increase[5-7] in dM/dh at some apparent h_c suggests that the free boson model is fairly good (i.e., the interaction effects are quite small).

III. MEAN-FIELD RESULTS

In the large-s approximation, the spin chain is equivalent to a quantum field theory, the $O(3)$ nonlinear σ model with Hamiltonian:

$$H=(v/2)\int dx[gl^2+(v/g)(\partial\boldsymbol{\varphi}/\partial x)^2]\ . \tag{3.1}$$

Here the field $\boldsymbol{\varphi}$ is constrained to have unit magnitude, $\boldsymbol{\varphi}^2=1$ and

$$l=(1/vg)\boldsymbol{\varphi}\times(\partial\boldsymbol{\varphi}/\partial t)\ . \tag{3.2}$$

The coupling constant, g, and spin-wave velocity takes the value

$$g=2/s,\ v=2Js\ . \tag{3.3}$$

The corresponding Lagrangian density is simply

$$L=(\tfrac{1}{2}g)[(1/v)(\partial\boldsymbol{\varphi}/\partial t)^2-v(\partial\boldsymbol{\varphi}/\partial x)^2]\ . \tag{3.4}$$

Due to the constraint, $\boldsymbol{\varphi}^2=1$, this is a highly nonlinear theory. There is a conserved rotation symmetry in the field theory; the associated conserved spin operators, obeying the SU(2) algebra are

$$S=\int dxl\ . \tag{3.5}$$

Thus $\boldsymbol{l}$ is the density of the conserved spin in the field theory. The original spin operators are expressed in terms of the field $\boldsymbol{\varphi}$ and the spin density, $\mathbf{l}$ as

$$\mathbf{S}_i\approx(-1)^i s\boldsymbol{\varphi}+\boldsymbol{l}\ . \tag{3.6}$$

i.e., $\boldsymbol{\varphi}$ is the sublattice magnetization density (the local Néel order parameter) and $\boldsymbol{l}$ is the uniform magnetization density (whose integral is conserved). The leading effect of the anisotropic term on the Hamiltonian density, H is

$$\delta H=Ds^2(\varphi^z)^2\ . \tag{3.7}$$

A magnetic field adds the term

$$\delta H=-g\mu_{Bi}hl^i\ , \tag{3.8}$$

i.e., the field couples to the conserved spin operators, $\mathbf{S}$. This theory is difficult to work with but a few results are known reliably. The unique ground state, $|0\rangle$, is a state of total spin zero; the excitation spectrum consists of a massive triplet, with the quantum numbers of

$$|i\rangle\propto\varphi^i|0\rangle\ ; \tag{3.9}$$

there are no bound states but repulsive interactions between the particles.

A much simpler theory that has all these features and arises in some circumstances from a renormalization group transformation[8] is the linear version of the model where the constraint is relaxed and a repulsive $\boldsymbol{\varphi}^4$ interaction is added,

$$H=(\tfrac{1}{2}v)(\partial\boldsymbol{\varphi}/\partial t)^2+(v/2)(\partial\boldsymbol{\varphi}/\partial x)^2+(\Delta^2/2v)\boldsymbol{\varphi}^2+\lambda\boldsymbol{\varphi}^4 . \tag{3.10}$$

Δ is the Haldane gap, which is inserted here as a phenomenological parameter and the λ term gives a repulsive interaction between the particles. A simple mean-field theory is then obtained by assuming λ is small and using the noninteracting theory (or low-order perturbation theory in λ if necessary). The free theory contains a triplet of bosons with a Lorentz invariant spectrum and rest energy Δ. The linear model also has a conserved spin operator with density:

$$\boldsymbol{l}=(1/v)\boldsymbol{\varphi}\times(\partial\boldsymbol{\varphi}/\partial t) . \tag{3.11}$$

The lattice spin operators are represented essentially the same way (up to a rescaling of the field):

$$\mathbf{S}_i\approx\sqrt{s}(-1)^i\boldsymbol{\varphi}+\boldsymbol{l} . \tag{3.12}$$

Note that because of the factor of $(-1)^i$, the actual momentum of the $\boldsymbol{\varphi}$ quanta is shifted by π. The spin structure function at k near π is given by the $\boldsymbol{\varphi}$ structure function, while the spin structure function near $k=0$ is given by the $\boldsymbol{l}$ structure function, which is a two-magnon operator. We phenomenologically model the anisotropy term by adjusting the gaps for the three modes to Δ_z and $\Delta_\pm$:

$$H=(\tfrac{1}{2}v)(\partial\boldsymbol{\varphi}/\partial t)^2+(v/2)(\partial\boldsymbol{\varphi}/\partial x)^2+(\Delta_z^2/2v)(\varphi^z)^2+(\Delta_\pm^2/2v)[(\varphi^x)^2+(\varphi^y)^2]+\lambda\boldsymbol{\varphi}^4 . \tag{3.13}$$

(The dependence of the gaps on D can be calculated in the large-n approximation. This will be reported elsewhere.) As before, the magnetic field adds the term:

$$\delta H=-g_a\mu_B h l^a . \tag{3.14}$$

All the desired properties of the spin chain can now be readily calculated using the free field approximation ($\lambda=0$). We use the standard mode expansion of φ^i in annihilation and creation operators:

$$\varphi^j(x,t)=\sum_k\sqrt{(v/4\pi L\omega_j)}\times\{\exp[-i(\omega_j t-kx)]a_{jk}+\text{H.c.}\} , \tag{3.15}$$

where a_{jk} is an annihilation operator for wave vector k and spin direction j,

$$\omega_j(k)^2=v^2k^2+\Delta_j^2 , \tag{3.16}$$

H.c. denotes Hermitian conjugate and L is the number of spins. For zero external field the Hamiltonian becomes

$$H=\sum_{ki}\omega_{ki}(a_{ki}^\dagger a_{ki}+\tfrac{1}{2}) . \tag{3.17}$$

The conserved total spin operator become:

$$S^z=i\sum_k(a_{kx}^\dagger a_{ky}-a_{ky}^\dagger a_{kx})=\sum_k(a_{k+}^\dagger a_{k+}-a_{k-}^\dagger a_{k-}) , \tag{3.18}$$

where we have used the linear combinations:

$$a_\pm\equiv(a_x\pm ia_y)/\sqrt{2} . \tag{3.19}$$

Thus the energies of the planar modes get shifted to:

$$\omega_\pm\to(v^2k^2+\Delta^2)^{1/2}\pm g_z\mu_B h , \tag{3.20}$$

while the z mode is unaffected as expected from general principles. The susceptibility per spin is

$$\chi^z(T)=(1/T)(g_z\mu_B)^2 2\int(dk/2\pi)n(k)[1+n(k)] , \tag{3.21}$$

where $n(k)$ is the boson occupation number:

$$n(k)\equiv1/[\exp(\omega_{k\pm}/T)-1] . \tag{3.22}$$

This has the expected (exponential) form of Eq. (2.14) at small T. At T large compared to Δ but small compared to the bandwidth, J, it becomes linear:

$$\chi^z(T)\to T/\Delta_\pm . \tag{3.23}$$

The magnetization has the general behavior discussed in Sec. II. It is zero at $T=0$ for $h<\Delta_\pm/g_z\mu_B$. In the free boson model it becomes infinite at h_c since an infinite number of bosons condense into the $k=0$ state. The effect of the repulsive interactions is to keep M (i.e., the boson number) finite.

As anticipated, the effect of a field in the x direction is considerably more complicated. The total spin operator (which is now not conserved because of the anisotropy term) is:

$$S^x=(i/2)\sum_k[(\sqrt{\omega_y/\omega_z}+\sqrt{\omega_z/\omega_y})(a_{ky}^\dagger a_{kz}-a_{kz}^\dagger a_{ky})+(\sqrt{\omega_y/\omega_z}-\sqrt{\omega_z/\omega_y})(a_{yk}a_{z,-k}-a_{yk}^\dagger a_{z,-k})] . \tag{3.24}$$

Note that the second term (only present for unequal gaps) can excite a pair of bosons from the vacuum and so it is active even at $T=0$. The susceptibility now becomes:

$$\chi^x(T)=g_\pm\mu_B)^2(\tfrac{1}{2})\int(dk/2\pi)[(1+n_y+n_z)(\omega_y-\omega_z)^2/(\omega_y+\omega_z)+(n_y-n_z)(\omega_y+\omega_z)^2/(\omega_z-\omega_y)]/\omega_y\omega_z . \tag{3.25}$$

There is now a nonzero susceptibility at $T=0$, proportional to the square of the difference of gaps:

$$\chi^x(0)=(g_x\mu_B)^2(\tfrac{1}{2})\int(dk/2\pi)(\omega_y-\omega_z)^2/[(\omega_y+\omega_z)\omega_y\omega_z] . \tag{3.26}$$

The leading T dependence at low T comes from the factor n_y and is hence $O(e^{-\Delta_\pm/T})$ as expected. At $\Delta \ll T \ll J$, χ is again linear in T:

$$\chi^x(T) \to (g_x\mu_B)^2 T(1/\Delta_\pm + 1/\Delta_z)/2 \ . \tag{3.27}$$

Note that although $\chi^z(0) < \chi^x(0)$, $\chi^z(T)$ has a larger slope in the intermediate temperature range; thus we might expect the two susceptibilities to cross at some T of order Δ.

A graph of the theoretical susceptibilities versus the experimental data from Ref. 4 is shown in Fig. 1. Note that once we have extracted the gaps and the velocity from the neutron-scattering data there are no free parameters in the susceptibility formulas; even the overall scale is completely determined. The theoretical curves seem to capture many of the features of the experimental data. The crossing of the two curves occurs at exactly the right place, for example. The main deficiency is that the experimental values of χ^z at low T are too high to fit the theory. This may be because of anisotropy terms in H, not included in this theory, which break the symmetry of rotation about the z axis. Indeed, the splitting of the measured values of χ^a and χ^c in Fig. 2 suggests the presence of some anisotropy. Alternatively, as discussed in Sec. II, the nonzero $\chi^b(0)$ may be due to extraneous effects. In any event there is no reason to expect better than qualitative agreement given the crude nature of the free boson approximation.

We may also calculate the effect of a finite field h_x on the dispersion relations. Note that in the free boson approximation the field in the x direction only mixes the y and z modes. Thus there is no change in the energy of the x mode. Again, this situation should change when the interaction effects are included. (There are Feynman diagrams for the x-self energy involving virtual loops of y and z particles.) To obtain the dispersion relation in the noninteracting approximation, we may express the complete Hamiltonian in terms of creation operators using (3.14), (3.17), and (3.24) and then diagonalize by a generalized Bogliubov transformation. A simpler procedure is to solve for the frequencies of the equivalent classical

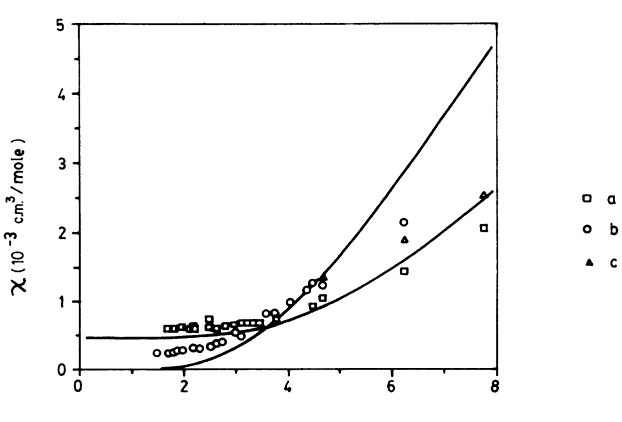

FIG. 1. Measured susceptibilities (Ref. 4) vs the predictions of the free boson model, Eqs. (3.21) and (3.25).

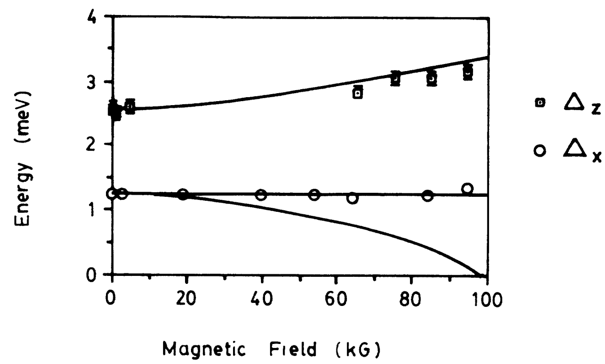

FIG. 2. Field-dependent gaps as determined by neutron scattering (Ref. 5) compared with predictions of the free boson model, Eq. (3.28).

problem. For a given value of wave vector, the corresponding harmonic oscillator Hamiltonian is

$$H = (\tfrac{1}{2}) \sum_{i=1}^{2} (\Pi_i^2 + \omega_i^2 \varphi_i^2) - g_x\mu_B h(\varphi_1\Pi_2 - \varphi_2\Pi_1) \ . \tag{3.28}$$

Here 1 and 2 refer to y and z modes. From the classical equations of motion:

$$d\varphi_i/dt = \partial H/\partial \Pi_i, d\Pi_i/dt = -\partial H/\partial \varphi_i \ , \tag{3.29}$$

we obtain the frequencies:

$$\omega_{z,y}(h)^2 = (\omega_z^2 + \omega_y^2)/2 + (g_x\mu_B h)^2 \pm [2(g_x\mu_B h)^2(\omega_z^2 + \omega_y^2) + (\omega_z^2 - \omega_y^2)^2/4]^{1/2} \ , \tag{3.30}$$

where on the right-hand side the energies are evaluated at $h = 0$. In the free boson model ω_y goes to zero at a critical field which has the same value as for a field in the z direction (up to the slight difference in g factors),

$$h_c = \Delta_\pm/(g_\pm \mu_B) \ . \tag{3.31}$$

However, unlike the case of a field in the z direction where the existence of the phase transition follows from symmetry arguments and general principles, it seems unlikely that the phase transition survives the effect of the interactions for a field in the x direction. However, if the boson repulsion is relatively weak [the effective coupling constant, λ in Eq. (3.10) is small] there may be a rather steep rise in dM^x/dh at approximately this value of h, i.e., an "effective h_{cx}." The combination of the nonzero interaction and the fact that $\Delta_z > \Delta_y$ should tend to push the effective h_{cx} somewhat higher than h_{cz}. The observed h_{cx} in Ref. 7 was about 33% higher.

The field-dependent gaps of Eq. (3.30) are compared with those obtained from neutron scattering in Fig. 2. Note that only two branches were observed in neutron scattering, whereas there should be three on general theoretical grounds. We suggest that the lower branch seen was actually the x branch [i.e., involving fluctuation of the spins in the direction of the applied field which was the c direction in the notation of Ref. 5]. The lowest y branch was not reported in Ref. 5. However, it was found that the intensity of the observed lower branch dropped by a factor of 2 as the field was increased to h_c. This could be explained if half the intensity split off into a lowest branch.

IV. CONCLUSIONS AND DIRECTIONS FOR FUTURE STUDY

All of the experimental measurements seem to be at least qualitatively consistent with the theory discussed here. Two apparent discrepancies that might be cleared up by further experimental work are the nonzero value of χ^b at low temperatures and the nonobservation of the splitting of x and y modes due to a field in the x direction, or due to anistropy even at zero field, if it exists.

More finite chain numerical work would also clearly be desirable. Calculations of the gaps Δ_z and $\Delta_\pm$ with the isotropy breaking D term of Eq. (2.1) could be used to estimate the value of D. Some work of this type has already been published[10] but the chains used were very short (mainly ten sites) and the gap estimated for $D=0$ of $0.25J$ is far off the present best estimates of about $0.4J$. It would also be useful to study the splitting of Δ_x and Δ_y modes with the addition of further anistropy such as a term:

$$\delta H = D_x \sum_i (S_i^x)^2 \ . \tag{4.1}$$

$T=0$ susceptibilities as a function of D and D_x would also be very useful and could help to clear up the question of the breaking of symmetry of rotation about the z axis in NENP. The $T=0$ magnetization has been studies numerically for the isotropic model in Ref. 11 and agrees well with the experimental results.[5-7] It would be interesting to extend these studies to lower fields and also to include anisotropy, the D term of Eq. (2.1). In particular the conjecture made here that there is no discontinuity in dM_x/dh_x for finite anisotropy could be checked. The free boson model did not agree very well with experiments on the apparent h_{cx} or the field-dependent gaps of Fig. 2. Numerical studies of these quantities at non-zero anisotropy parameter, D, would also be useful.

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