Quantum Antiferromagnets in a Magnetic Field

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Motivated by recent experiments on low-dimensional quantum magnets in applied magnetic fields, we present a theoretical analysis of their properties based on the nonlinear \( \sigma \) model. The spin stiffness and a 1/N expansion are used to map the regimes of spin-gap behavior, predominantly linear magnetization, and spin saturation. A two-parameter renormalization-group study gives the characteristic properties over the entire parameter range. The model is relevant to many systems exhibiting Haldane physics, and is applied here to the two-chain spin ladder compound CuHpCl.

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The importance of low-dimensional spin systems in revealing fundamentally new quantum mechanical properties has been recognized since Haldane’s conjecture concerning the effects of quantum fluctuations in integer- and half-integer-spin antiferromagnetic (AF) chains. We begin with the experimental observation that the magnetization curves of some such materials, thought to be prototypical of the extreme quantum limit, are in fact remarkably classical. As examples, we refer here to the Haldane (\( S = 1 \)) chain NENP, and primarily to the two-chain CuHpCl, which show regimes of linear magnetization whose gradient is the (classical) Néel susceptibility.

With the goal of understanding such behavior, we consider the quantum AF system in an external magnetic field using the nonlinear \( \sigma \) model (NLsM). This widely-applied treatment is in fact semi-classical, being truly valid only in the limit of large spin \( S \), but has in the past formed the basis for many fundamental deductions concerning the quantum limit. We will demonstrate here its applicability for effectively integer-spin quantum systems in appreciable magnetic fields, and provide justification of this result in terms of the suppression of quantum fluctuations by the field. For the purposes of developing the present theoretical analysis, we will concentrate on the best-characterized sample in recent literature, Cu$_2$(1,4-diazacycloheptane)$_2$Cl$_4$ (CuHpCl).

The Hamiltonian for the ladder system in a magnetic field \( b = \pm \mu_B B \) may be written as

\[
H = \sum_{i,m=1,2} [J S_{m,i} \cdot S_{m,i+1} + J' S_{1,i} \cdot S_{2,i} + b \cdot S_{m,i}],
\]

where \( J \) is the intrachain exchange interaction and \( J' \) the interchain, or ladder “rung,” interaction. The derivation of the NLsM in the presence of a magnetic field is presented in Ref. [1]. For \( N_x \)-site chains with periodic boundary conditions along \( x \), in the geometry shown in Fig. 1, the resulting model has \( J_x = J \), \( J_y = \frac{1}{2} J' \), \( b = (0,0,b) \), and \( S_{m,N_x+i} = S_{m,i} \). There are two key points. First, the spin is written in terms of slowly-varying, orthogonal, staggered and uniform components as \( S_{m,i} = S((-1)^{i+m} n_{m,i} + a l_{m,i}) \), where \( a \) is the lattice constant, and the fluctuations \( I \) about the staggered configuration must be integrated out subject to the orthogonality constraint \( n \cdot 1 = 0 \). Second, the full Euclidean action in space and inverse temperature, \( S_E = S_B + \int_0^\beta d\tau H \), contains in addition a Berry-phase term \( S_B = i \frac{\pi}{2} \int d\tau dx [-aI(n \wedge \dot{n}) + 4\pi i S(F_1 + P_2) \frac{P_1}{P_2}]. \frac{P_1}{P_2} = \frac{1}{2\pi} \int d\tau dx (n \wedge \dot{n}) \cdot \partial_x n \) separate to give simply the Pontryagin index on each chain when \( \partial_x n = 0 \), as is the case in a ladder of only two chains. The last term in \( S_B \) is thus \( i(4\pi P)2S \), demonstrating that the system will have integer-spin characteristics for any value of \( S \), and the topological term may be ignored.

The action for the quasi-one-dimensional ladder system, in 1+1 Euclidean dimensions denoted by \( \mu \), is then

\[
S_E = \frac{1}{2g} \int_{\tau,x} [(|\partial_\mu n|^2 - (b^2 - (n \cdot b)^2) + 2ib \cdot n \wedge \dot{n}].
\]

where \( g = (2/N_y) \sqrt{J/J_z} \) is the bare coupling constant, \( J = J_x + \frac{1}{2} J_y \), and the integral over \( \tau \) is to upper limit \( L_T = c\beta \), with \( c = 2S a \sqrt{J_z/J}/h \) \((h \to 1)\) the effective spin-wave velocity. We have left explicit the number \( N_y \) of chains in the ladder; for the compound under consideration \( J_y > J_z \) and \( N_y = 2 \), giving an effectively rigid rung coupling. The form of the NLsM in an external field given by the second term in Eq. (4) has been derived previously, and its implications considered in the low-field limit. In what follows we will examine its effects for arbitrary fields.

To gain initial insight into the effect of the magnetic field, we consider the spin stiffness of the ladder system. Taking the staggered spin configuration to be subject to a twist \( \theta \) in the plane normal to the applied field (Fig. 1), we calculate the free energy \( F(b,\theta) \) to one-loop order in the important out-of-plane spin fluctuations, and deduce the spin stiffness from \( \rho_s = \frac{1}{2c} \frac{L}{V} \frac{\partial^2 F}{\partial \theta^2} |_{\theta = 0} \) .

\[
\rho_s = \rho_s^0 \left[ 1 - \frac{g}{L_T} \sum_k \frac{1}{k^2 + (b/c)^2} \right],
\]

where \( \rho_s^0 = c/2g \) is the classical value and the sum
provides both quantum and thermal (through the finite “length”) corrections to first order.

We restrict the analysis to the low-temperature, or “quantum” case \( L_T > L \). Evaluating the summation between spatial limits \( \pi/L \) and \( \pi/a \), and introducing the “magnetic length” \( L_m \) as \( \pi/L_m = b/c \),

\[
\rho_s = \rho_s^0 \left[ 1 + \frac{g}{4\pi} \ln \left( \frac{(a/L)^2 + (a/L_m)^2}{1 + (a/L_m)^2} \right) \right].
\]

\( L \) in Eq. (4) may be regarded as the correlation length \( \xi \), beyond which segments of the ladder behave independently, and can be computed from \( \rho_s = 0 \). In the weak-field limit \((L_m \to \infty)\) one recovers the result \( \xi_0 = ae^{2\pi/2} = ae^{\alpha\pi S} \), where \( \alpha = \sqrt{Jz/J} \). The general solution is \( \xi(B) = \xi_0/\sqrt{1-(L_m/L_m)^2} \), where \( L_m = a/e^{2\pi/2} - 1 \) gives the critical field \( B^* \) at which the correlation length diverges. For fields \( B < B^* \), the finite correlation length may be written as \( \xi(B) = ae^{\alpha\pi S} \), where \( S = S(1-g/4\pi \ln(1-(L_m/L_m)^2)) \) is a growing value of the effective spin. For \( B > B^* \), the field enforces a quasi-long-correlation throughout the system \( S \), and it is most convenient to write the spin stiffness as \( \rho_s = \rho_s^0[1-g/4\pi \ln(1 + (L_m/a)^2)] \), which recovers the bare value as \( B \to \infty \). Finally, the divergence of the correlation length at \( B^* \) corresponds to the closing of the gap \( \Delta \) to spin excitations according to \( \Delta \propto \sqrt{1-(B/B^*)^2} \). This situation is summarized in Fig. 2.

However, if the \( B \) derivative is taken of the free energy in order to compute the magnetization, the resulting terms do not yield the required zero result in the spin gap regime. This indicates the breakdown of the approximation implicit in deriving \( F(b, \theta) \) that the field be large on the scale of in-plane fluctuation energies \( |\phi| \). Thus the spin stiffness analysis, while qualitatively revealing of the behavior of the system, is not reliable at low fields.

The situation in the weak-field regime may be addressed by a \( 1/N \) expansion [14], which is expected to be appropriate in describing spin-gap phases. The staggered spin \( \mathbf{n} \) is taken to exist in an \( N \)-dimensional spin space, in which only the component \( n_z \) is selected by the field. The relevant parts of the \( O(N) \) action are

\[
S_E = \frac{1}{2g} \int d\mathbf{x} \left( \partial_\mu \mathbf{n} \right)^2 - \vec{b}^2(1-n_z^2) - i\lambda(n^2-1),
\]

where \( \vec{b} \) denotes \( b/c \) and the constraint that \( n \) have unit magnitude is made explicit with the Lagrange multiplier \( i\lambda \), whose saddle-point value is given by

\[
\frac{1}{g} = (N-1) \sum_k \frac{1}{k^2 + i\lambda} + \sum_k \frac{1}{k^2 + i\lambda + b^2},
\]

in which the \( B \)-field term is found to appear only at \( O(1/N) \). \( i\lambda \) functions as a mass, or cutoff term in momentum integrations, and is thus an upper lengthscale for cooperative processes in the system, or simply a correlation length (inverse excitation gap). Writing the saddle-point solution as \( i\lambda = e^2\pi^2/\xi(B)^2 \) and carrying out the summation at low \( T \) gives an expression analogous to \( \rho_s = 0 \) emerging from Eq. (4).

At weak fields \( (\xi \ll L_m) \), one finds \( \xi = a/e^{2\pi/2}(1 + (a^2/NL_m^2)e^{4\pi/2}) \), a \( 1(1/N) \) correction to a result differing from the previous one by a power of \( 1/N \) in the exponent. The result \( \frac{\partial}{\partial b} \left( \frac{e^{2\pi/2}}{\xi} \right) = -\frac{2b}{N} \) ensures both that the corresponding magnetization contribution from the \( k \) summation terms in \( F \) is identically zero to \( O(1/N) \), and that the constant term \( b^2 + e^2\pi^2/\xi^2 \) yields \( 2b(1-1/N) \). It is clear that the behavior required of a gapped system is returned in the weak-field regime only on making the well-recognized identification (motivated by comparison with renormalization-group results [14]) \( N \to N-2 \), and by returning to the physical situation \( N = 3 \). Then the magnetization is indeed zero, and the saddle-point solution for \( \xi(B) \) becomes precisely that deduced from the spin stiffness analysis, with the same critical field \( B^* \). With this replacement caveat we thus obtain from these two approaches a consistent picture of both weak- and strong-field regimes.

To quantify the regimes of validity of the foregoing analyses, we consider next a renormalization-group (RG) approach to the NLsM in an applied field. Taking the model in the form

\[
S_E = \frac{1}{2g} \int d\mathbf{x} \left( \partial_\mu \mathbf{n} \right)^2 - \vec{b}^2(1-n_z^2) + 4i\vec{n}_x \dot{n}_y
\]

and transforming to variables \( \phi \) and \( \sqrt{2}\sigma_z = n_z \) representing respectively the in- and out-of-plane fluctuations, the latter in a form suitable for perturbative expansion in the coupling constant \( g \), we obtain

\[
\mathcal{L}_E = \frac{1}{2g} \left( A - \vec{b}^2 \right) - \frac{1}{2} \sigma_z \left( -\partial_\mu^2 \vec{b}^2 - A \right) \sigma_z + O(g).
\]
$A$ denotes the in-plane terms $(\partial_x \phi)^2 + 2i\bar{\phi}$, which because $\phi(\tau, x)$ is assumed to vary slowly can be taken to be a small constant (no fast Fourier modes) in the momentum shell $|\pi/a < |k| < \pi/a$ ($\gamma \to 1$). Performing the integral and expanding in $A$, the form of Eq. (6) is recovered with coefficients $g(a')$ and $b(a')^2$ given by partial traces. Evaluation of these and differentiation with respect to the flow parameter $l = \ln(a'/a)$, leads to the coupled RG equations

$$\frac{dg}{dl} = \frac{g^2}{2\pi} \frac{1}{1 + \beta^2}, \quad \frac{d\beta^2}{dl} = 2\beta^2 - \frac{g^2}{2\pi} \ln \left(1 + \beta^2\right), \quad (9)$$

in which $\beta = a^2b(a')$. These new RG equations possess a variety of interesting limiting cases, whose detailed study we defer to a future publication [13].

For the present purposes, we concentrate on the fixed points to obtain a qualitative picture of the RG flow diagram, and on the consequences for the magnetization. (i) Seeking a fixed point by weak-field expansion around $\beta_0 = 0$, we find $dg/dl \simeq g^2/2\pi$ and thus $d\ln \beta^2/dl = 2 - d\ln g/dl$, which may be solved to yield

$$\frac{g_0}{g} = 1 - \frac{g_0}{2\pi} l, \quad \beta = \beta_0 e^{\left[1 - \frac{g_0}{2\pi}\right]^{1/2}}. \quad (10)$$

The fixed point $(g_*, \beta_*) = (\infty, 0)$ is clearly stable if the flow is stopped at $l_* = 2\pi/g_0$. The system will flow to this strong-coupling regime if the starting value $\beta_0$ is sufficiently small. The lengthscale $L_* = ae^{\pi/\beta_0}$ at which the flow stops may be compared with the spatial and thermal dimensions $L, L_T$ of the system to calculate directly the effects of finite size and temperature [13]. (ii) At strong fields ($\beta \to \infty$), $dg/dl = 0$, or $g = g_0$, indicating that the coupling is not renormalized, and $d\ln \beta^2 = 2dl$, from which it follows that $b(l) = \beta_0$, i.e. neither is the field.

Numerical solution of Eqs. (6) leads to the flow diagram in Fig. 3. The regime (ii) of strong initial field may be termed the weak-coupling situation, where $g$ and $b$ are weakly renormalized to finite values. Here the perturbation theoretic approach is consistent and the weak coupling corresponds to deconfinement of the excitations on the lengthscale $L_m$ set by the field. The regime (i) is the strong-coupling condition, of confinement of (gapped) excitations, where the assumption of small $g$ in the derivation proves to be inconsistent. However, in this regime one may deduce the critical lengthscale $L_*$ governing the behavior of the system and, as we will show below, that the magnetization is zero (inset Fig. 3). The critical starting field separating the two regions is $\tilde{b}_* = 0.46$. The properties of the two regimes may be illustrated by considering the correlation length in each. $\xi$ is a physical quantity and does not change under the RG flow, meaning that $d\xi/d\beta = 0$, whence

$$\frac{\partial g}{\partial l} \frac{\partial \xi}{\partial g} + \frac{\partial \beta^2}{\partial l} \frac{\partial \xi}{\partial \beta^2} + \xi = 0. \quad (11)$$

(i) At small $\beta^2$, $-\partial \xi/\xi \simeq 2\pi g/\mu_0$, which has solution $\xi_0 = \mu_0 e^{2\pi\xi/\xi_0}$ (see above) for the finite physical correlation length. (ii) For large $\beta^2$, $-2\partial \xi/\xi \simeq \partial \beta^2/\beta^2$ leads to $\xi_0 = \xi L/a (b_0$ invariant) under the RG flow, and we see that for any assumed finite $\xi$, the bare correlation length $\xi_0$ is the system size $L$ or $L_T$.

We return to the experimental motivation for the above analysis, and compute the magnetization $M = \partial F/\partial B$ of the model over the full field range,

$$M = -g_0B N_x N_y (b/4J) + M_d + M_f. \quad (12)$$

$M$ contains contributions linear in $B$ from the quadratic term $\bar{B}$, $M_d$ from the dynamical term in the trace (cf. $\bar{B}$) due to fluctuations of $n$ out of the plane perpendicular to $B$, and $M_f$ from in-plane fluctuation terms $\phi$. The last give a sawtooth form leading to magnetization steps [13], a finite-size effect which will not be considered further here. While the linear term is always present, we have shown that below a threshold field $B^*$, where the system has a spin gap, it is cancelled by the corresponding correlation-length term. Above an upper threshold $B_{\text{us}}$, the magnetization will saturate at the value $M_s = g_0B S N_x (N_x = N_y g_0)$, and this effect is not contained in the (large-S) model as applied. Systematic inclusion of an additional total spin constraint is possible [14], but to compress the analysis we will here apply $M_s$ as a simple cutoff. The “dynamical” contribution is given consistently to lowest order in the small parameter $c/b$, and in limit of low $T$, by the constant $M_d = \frac{1}{2} N_x g_0 B$. Next-order corrections involving the spin excitations have the form $M_d' \sim B \ln(B/J)$.

Specializing to the two-chain ladder material CuHpCl, the exchange constants deduced from the magnetization and susceptibility [3] are $J' = 13.2K$ and $J = 2.4K$, whence $J = 13.3T$ and $J = 3.6T$. Taking the simplest case of constant $M_d$, and the lower critical field $B_{\text{sc}}$ for

![FIG. 3. RG flow diagram for $g$ and $\tilde{b}$. Strong- and weak-coupling regimes are separated by separatrix $s$.](image-url)
onset of magnetization where \( M(B_{c2}) = 0 \), we obtain \( B_{c1} = J/\bar{g} = 6.6T \). The saturation field \( B_{c2} \) is given from \( M(B_{c2}) = M_s \) as \( B_{c2} = 45/\bar{g}J = 13.3T \). These values are in remarkably good agreement with a linear extrapolation of the magnetization data at lowest temperature in Ref. [3], where \( B_{c1} = 6.8T \) and \( B_{c2} = 13.7T \). Such an extrapolation appears closer to the data than the predictions \(^4\) of a repulsive boson model. We note further that the gradient of the linear magnetization is precisely the classical Néel susceptibility \( \chi_{AF} = (\bar{g}m_B)^2/(4J\bar{a}^2) \) per unit volume, a result in itself remarkable for an AF in the quantum limit. The computed magnetization is shown in Fig. 4, where the dashed line indicates the validity limit of the calculation.

\[ M = \frac{\bar{g}m_B}{c} \frac{\partial F}{\partial \beta_0} = \frac{\bar{g}m_B}{c} \left( \frac{\partial \beta}{\partial \beta_0} \frac{\partial F}{\partial \beta} + \frac{\partial \phi}{\partial \beta_0} \frac{\partial F}{\partial \phi} \right), \]

from which we see in the small-B regime, where \( \partial \phi/\partial \beta_0 = 0 \) and \( \partial \beta/\partial \beta_0 = c^l(1 - g\phi/2\pi) \), that for any \( F(\beta) \) analytic in \( \beta \), \( M(B_0) \sim \sqrt{1 - g\phi/2\pi \ln(L/a)} \to 0 \) as \( L \to L^\ast \). Thus the magnetization vanishes in the strong-coupling limit as required. From the numerical solution, the field scale \( b \), gives \( B_s = 1.6T \), in reasonable agreement with the value \( B^\ast \) above. We have already shown that the regime approaching the physical \( B_{c1} \) lacks a suitable calculation scheme. Physically, the problem is one of determining the effects of quantum fluctuations of the spin system when the spin gap is weakened by the applied field, and the quantum disordered phase thus made less robust. We have detailed above the lowest-order predictions of the NLsM for the closing of the gap, and expect these to be appropriate when the gap becomes smaller than other energy scales (quantum, thermal and finite-size fluctuations) in the system.

The foregoing analysis is not restricted to CuHpCl, but applies also to the \( S = 1 \) AF ("Haldane") chain. NENP is considered to be a prototypical case for observation of the Haldane gap but for the complication of a large single-ion anisotropy. The present study predicts again the qualitative features of a gapped regime of zero magnetization, followed by approximately linear behaviour towards saturation (not achieved), as in experiment \(^2\). We will present elsewhere the quantitative aspect of this problem. There has been considerable recent interest in the possibility of magnetization plateaus in certain systems, and we observe that in the current model these may be expected, for example in \( S > 1 \) chains, when the field strength is such that the projected in-plane spin \( S_{\perp} \) is of integer amplitude, leading to a gapped phase.

In summary, the nonlinear \( \sigma \) model treatment reproduces well the behavior of quantum antiferromagnets in an external field. For effectively integer-spin systems, meaning those with a trivial topological term, this statement is valid even in the extreme quantum limit of low spin and low dimensionality, because the magnetic field acts to suppress quantum fluctuation effects.

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