Bond operator methods for dimerized antiferromagnets

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1 Coupled dimer antiferromagnet

We begin by describing the quantum phase transition in a simple two-dimensional model of antiferromagnetically coupled $S = 1/2$ Heisenberg spins which has 2 spins per unit cell. The transition is tuned by varying a dimensionless parameter $\lambda$. As we noted in Section ?? different ‘dimerized’ Mott insulators will correspond to different values of $\lambda$, and the value of $\lambda$ can be tuned by applying pressure [8, 10].

We consider the “coupled dimer” Hamiltonian [25]

$$H_d = J \sum_{(ij) \in A} \mathbf{S}_i \cdot \mathbf{S}_j + \lambda J \sum_{(ij) \in B} \mathbf{S}_i \cdot \mathbf{S}_j$$

where $\mathbf{S}_j$ are spin-1/2 operators on the sites of the coupled-ladder lattice shown in Fig[1] with the $A$ links forming decoupled dimers while the $B$ links couple the dimers as shown. The ground state of $H_d$ depends only on the dimensionless coupling $\lambda$, and we will describe the low temperature ($T$) properties as a function of $\lambda$. We will restrict our attention to $J > 0$ and $0 \leq \lambda \leq 1$.

Note that exactly at $\lambda = 1$, $H_d$ is identical to the square lattice antiferromagnet, and this is the only point at which the Hamiltonian has only one spin per unit cell. At all other values of $\lambda$ $H_d$ has a pair of $S = 1/2$ spins in each unit cell of the lattice. As will become clear from our discussion, this is a key characteristic which permits a simple theory for the quantum phase transition exhibited by $H_d$. Models with only a single $S = 1/2$ spin per unit cell usually display far more complicated behavior, and will be discussed in Sections ??,??.

We will begin with a physical discussion of the phases and excitations of the coupled dimer antiferromagnet, $H_d$ in Section [1] We will propose a quantum field-theoretical description of this model in Section [12] we will verify that the limiting regimes of the field theory contain excitations whose quantum numbers are in accord with the phases discussed in Section [1] and
will then use the field theory to describe the quantum critical behavior both at zero and finite temperatures.

1.1 Phases and their excitations

Let us first consider the case where \( \lambda \) is close to 1. Exactly at \( \lambda = 1 \), \( H_d \) is identical to the square lattice Heisenberg antiferromagnet, and this is known to have long-range, magnetic Néel order in its ground state, i.e. the spin-rotation symmetry is broken and the spins have a non-zero, staggered, expectation value in the ground state with

\[
\langle S_j \rangle = \eta_j N_0 n,
\]

where \( n \) is some fixed unit vector in spin space, \( \eta_j \) is \( \pm 1 \) on the two sublattices, and \( N_0 \) is the Néel order parameter. This long-range order is expected to be preserved for a finite range of \( \lambda \) close to 1. The low-lying excitations above the ground state consist of slow spatial deformations in the orientation \( n \): these are the familiar spin waves, and they can carry arbitrarily low energy, i.e. the phase is ‘gapless’. The spectrum of the spin waves can be obtained from a text-book analysis of small fluctuations about the ordered Néel state using the Holstein-Primakoff method \[26\]: such an analysis yields two polarizations of spin waves at each wavevector \( k = (k_x, k_y) \) (measured from the antiferromagnetic ordering wavevector), and they have excitation energy

\[
\varepsilon_k = (c_x^2 k_x^2 + c_y^2 k_y^2)^{1/2},
\]

with \( c_x, c_y \) the spin-wave velocities in the two spatial directions.

Let us turn now to the vicinity of \( \lambda = 0 \). Exactly at \( \lambda = 0 \), \( H_d \) is the Hamiltonian of a set of decoupled dimers, with the simple exact ground state wavefunction shown in Fig. 2: the spins in each dimer pair into valence bond singlets, leading to a paramagnetic state which preserves spin rotation invariance and all lattice symmetries. Excitations are now formed by breaking a valence bond, which leads to a three-fold degenerate state with total spin \( S = 1 \), as shown in Fig. 3a. At \( \lambda = 0 \), this broken bond is localized, but at finite \( \lambda \) it can hop from site-to-site, leading to a triplet quasiparticle excitation. Note that this quasiparticle is not a spin-wave (or equivalently, a ‘magnon’).
Fig. 2. Schematic of the quantum paramagnet ground state for small $\lambda$. The ovals represent singlet valence bond pairs.

Fig. 3. (a) Cartoon picture of the bosonic $S = 1$ excitation of the paramagnet. (b) Fission of the $S = 1$ excitation into two $S = 1/2$ spinons. The spinons are connected by a “string” of valence bonds (denoted by dashed ovals) which lie on weaker bonds; this string costs a finite energy per unit length and leads to the confinement of spinons.

but is more properly referred to as a spin 1 exciton or a triplon. We parameterize its energy at small wavevectors $k$ (measured from the minimum of the spectrum in the Brillouin zone) by

$$\varepsilon_k = \Delta + \frac{c_x^2 k_x^2 + c_y^2 k_y^2}{2\Delta},$$

(3)

where $\Delta$ is the spin gap, and $c_x, c_y$ are velocities; we will provide an explicit derivation of (3) in Section 1.2. Fig 3 also presents a simple argument which shows that the $S = 1$ exciton cannot fission into two $S = 1/2$ 'spinons'.

The very distinct symmetry signatures of the ground states and excitations between $\lambda \approx 1$ and $\lambda \approx 0$ make it clear that the two limits cannot be continuously connected. It is known that there is an intermediate second-order phase transition at $\lambda_c = 0.52337(3)$ between these states as shown in Fig 4. Both the spin gap $\Delta$ and the Néel order parameter $N_0$ vanish continuously as $\lambda_c$ is approached from either side.

1.2 Bond operators and quantum field theory

In this section we will develop a continuum description of the low energy excitations in the vicinity of the critical point postulated above. There are a
Spin gap paramagnet. S=1 triplet quasiparticle excitations

Neel order. Doublet spin-wave excitations

Fig. 4. Ground states of $H_d$ as a function of $\lambda$. The quantum critical point is at $\lambda_c = 0.52337(3)$. The compound TlCuCl$_3$ undergoes a similar quantum phase transition under applied pressure \cite{8}.

number of ways to obtain the same final theory: here we will use the method of bond operators \cite{14} \cite{15}, which has the advantage of making the connection to the lattice degrees of freedom most direct. We rewrite the Hamiltonian using bosonic operators which reside on the centers of the $A$ links so that it is explicitly diagonal at $\lambda = 0$. There are 4 states on each $A$ link ($|\uparrow\downarrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$, and $|\downarrow\downarrow\rangle$) and we associate these with the canonical singlet boson $s$ and the canonical triplet bosons $t_\alpha (\alpha = x,y,z)$ so that

$$|s\rangle \equiv s^\dagger|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) ; \quad |t_x\rangle \equiv t_x^\dagger|0\rangle = \frac{-1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) ;$$

$$|t_y\rangle \equiv t_y^\dagger|0\rangle = \frac{i}{\sqrt{2}}(|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle) ; \quad |t_z\rangle \equiv t_z^\dagger|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle). \quad (4)$$

Here $|0\rangle$ is some reference vacuum state which does not correspond to a physical state of the spin system. The physical states always have a single bond boson and so satisfy the constraint

$$s^\dagger s + t_\alpha^\dagger t_\alpha = 1 \quad (5)$$

By considering the various matrix elements $\langle s|S_1|t_\alpha\rangle$, $\langle s|S_2|t_\alpha\rangle$, ..., of the spin operators $S_1,2$ on the ends of the link, it follows that the action of $S_1$ and $S_2$ on the singlet and triplet states is equivalent to the operator identities

$$S_{1\alpha} = \frac{1}{2} \left( s^\dagger t_\alpha + t_\alpha^\dagger s - i\epsilon_{\alpha\beta\gamma} t_\beta^\dagger t_\gamma \right) ,$$

$$S_{2\alpha} = \frac{1}{2} \left( -s^\dagger t_\alpha - t_\alpha^\dagger s - i\epsilon_{\alpha\beta\gamma} t_\beta^\dagger t_\gamma \right) . \quad (6)$$
where $\alpha, \beta, \gamma$ take the values $x, y, z$, repeated indices are summed over and $\epsilon$ is the totally antisymmetric tensor. Inserting (6) into (11), and using (5), we find the following Hamiltonian for the bond bosons:

\[
H_d = H_0 + H_1
\]

\[
H_0 = J \sum_{\ell \in A} \left( -\frac{3}{4} s_{\ell}^1 s_{\ell}^1 + \frac{1}{4} t_{\ell \alpha}^1 t_{\ell \alpha}^1 \right)
\]

\[
H_1 = \lambda J \sum_{\ell, m \in A} \left[ a(\ell, m) \left( t_{\ell \alpha}^1 t_{m \alpha}^1 s_{m}^1 s_{\ell}^1 + t_{\ell \alpha}^1 t_{m \alpha}^1 s_{m} s_{\ell} + \text{H.c.} \right) \right. + b(\ell, m)
\]

\[
\times \left( i^{\alpha \beta \gamma} t_{m \alpha}^1 t_{\ell \beta}^1 t_{\ell \gamma}^1 s_{m} + \text{H.c.} \right) + c(\ell, m) \left( t_{\ell \alpha}^1 t_{m \beta}^1 t_{\ell \beta}^1 - t_{\ell \alpha}^1 t_{m \beta}^1 t_{\ell \beta}^1 \right) \right]
\]

where $\ell, m$ label links in $A$, and $a, b, c$ are numbers associated with the lattice couplings which we will not write out explicitly. Note that $H_1 = 0$ at $\lambda = 0$, and so the spectrum of the paramagnetic state is fully and exactly determined. The main advantage of the present approach is that application of the standard methods of many body theory to (7), while imposing the constraint (5), gives a very satisfactory description of the phases with $\lambda \neq 0$, including across the transition to the Néel state. In particular, an important feature of the bond operator approach is that the simplest mean field theory already yields ground states and excitations with the correct quantum numbers; so a strong fluctuation analysis is not needed to capture the proper physics.

A complete numerical analysis of the properties of (7) in a self-consistent Hartree-Fock treatment of the four boson terms in $H_1$ has been presented in Ref. [14]. In all phases the $s$ boson is well condensed at zero momentum, and the important physics can be easily understood by examining the structure of the low energy action for the $t_\alpha$ bosons. For the particular Hamiltonian (1), the spectrum of the $t_\alpha$ bosons has a minimum at the momentum $(0, \pi)$, and for large enough $\lambda$ the $t_\alpha$ condense at this wavevector: the representation (6) shows that this condensed state is the expected Néel state, with the magnetic moment oscillating as in (2). The condensation transition of the $t_\alpha$ is therefore the quantum phase transition between the paramagnetic and Néel phases of the coupled dimer antiferromagnet. In the vicinity of this critical point, we can expand the $t_\alpha$ field in gradients away from the $(0, \pi)$ wavevector: so we parameterize

\[
t_{\ell, \alpha}(\tau) = t_\alpha(r_\ell, \tau)e^{i(0, \pi) \cdot r_\ell}
\]

where $\tau$ is imaginary time, $r \equiv (x, y)$ is a continuum spatial co-ordinate, and expand the effective action in spatial gradients. In this manner we obtain

\[
S_t = \int d^2r d\tau \left[ t_0 \frac{\partial t_\alpha}{\partial \tau} + C t_\alpha + \frac{D}{2} (t_\alpha t_\alpha + \text{H.c.}) + K_{1x} |\partial_x t_\alpha|^2 + K_{1y} |\partial_y t_\alpha|^2
\]

\[
+ \frac{1}{2} (K_{2x} (\partial_x t_\alpha)^2 + K_{2y} (\partial_y t_\alpha)^2 + \text{H.c.}) + \cdots \right].
\]
Here $C, D, K_{1,2,x,y}$ are constants that are determined by the solution of the self-consistent equations, and the ellipses represent terms quartic in the $t_\alpha$. The action $S_t$ can be easily diagonalized, and we obtain a $S = 1$ quasiparticle excitation with the spectrum

$$\varepsilon_k = \left[ (C + K_{1x} k_x^2 + K_{1y} k_y^2)^2 - (D + K_{2x} k_x^2 + K_{2y} k_y^2)^2 \right]^{1/2}. \quad (10)$$

This is, of course, the triplon (or spin exciton) excitation of the paramagnetic phase postulated earlier in (3); the latter result is obtained by expanding (10) in momenta, with $\Delta = \sqrt{C^2 - D^2}$. This value of $\Delta$ shows that the ground state is paramagnetic as long as $C > D$, and the quantum critical point to the Néel state is at $C = D$.

The critical point and the Néel state are more conveniently described by an alternative formulation of $S_t$ (although an analysis using bond operators directly is also possible [29]). It is useful to decompose the complex field $t_\alpha$ into its real and imaginary parts as follows

$$t_\alpha = Z(\varphi_\alpha + i\pi_\alpha), \quad (11)$$

where $Z$ is a normalization chosen below. Insertion of (11) into (9) shows that the field $\pi_\alpha$ has a quadratic term $\sim (C + D)\pi_\alpha^2$, and so the co-efficient of $\pi_\alpha^2$ remains large even as the spin gap $\Delta$ becomes small. Consequently, we can safely integrate $\pi_\alpha$ out, and the resulting action for $\varphi_\alpha$ takes the form

$$S_\varphi = \int d^2r d\tau \left\{ \frac{1}{2} \left( \partial_\tau \varphi_\alpha \right)^2 + c_x^2 (\partial_x \varphi_\alpha)^2 + c_y^2 (\partial_y \varphi_\alpha)^2 + s\varphi_\alpha^2 \right\} + \frac{u}{24} \left( \varphi_\alpha^2 \right)^2. \quad (12)$$

Here we have chosen $Z$ to fix the co-efficient of the temporal gradient term, and $s = C^2 - D^2$.

The formulation $S_\varphi$ makes it simple to explore the physics in the region $s < 0$. It is clear that the effective potential of $\varphi_\alpha$ has a minimum at a non-zero $\varphi_\alpha$, and that $\langle \varphi_\alpha \rangle \propto N_0$, the Néel order parameter in (2). It is simple to carry out a small fluctuation analysis about this saddle point, and we obtain the doublet of gapless spin-wave modes advertised earlier.

We close this subsection by noting that all of the above results have a direct generalization to other lattices, and also to spin systems in three dimensions. Matsumoto et al. [10] have applied the bond operator method to TlCuCl$_3$ and obtained good agreement with experimental observations. One important difference that emerges in such calculations on some frustrated lattices [30] is worth noting explicitly here: the minimum of the $t_\alpha$ spectrum need not be at special wavevector like $(0, \pi)$, but can be at a more generic wavevector $Q$ such that $Q$ and $-Q$ are not separated by a reciprocal lattice vector. A simple example which we consider here is an extension of (1) in which there are additional exchange interactions along all diagonal bonds oriented ‘north-east’ (so that the lattice has the connectivity of a triangular lattice). In such
cases, the structure of the low energy action is different, as is the nature of
the magnetically ordered state. The parameterization (8) must be replaced by
\[ t_{\ell\alpha}(\tau) = t_{1\alpha}(r_{\ell}, \tau)e^{iQ \cdot r_{\ell}} + t_{2\alpha}(r_{\ell}, \tau)e^{-iQ \cdot r_{\ell}} \] (13)
where \( t_{1,2\alpha} \) are independent complex fields. Proceeding as above, we find that
the low energy effective action (12) is replaced by
\[ S_{\Phi} = \int d^2rd\tau \left[ \frac{1}{2} \left( |\partial_{\tau}\Phi_{\alpha}|^2 + c_{\|}^2 |\partial_{x}\Phi_{\alpha}|^2 + c_{\perp}^2 |\partial_{y}\Phi_{\alpha}|^2 + s |\Phi_{\alpha}|^2 \right) + \frac{u}{2} \left( |\Phi_{\alpha}|^2 \right)^2 + \frac{v}{2} |\Phi_{\alpha}^2|^2 \right]. \] (14)
where now \( \Phi_{\alpha} \) is a complex field such that \( \langle \Phi_{\alpha} \rangle \sim \langle t_{1\alpha} \rangle \sim \langle t_{2\alpha} \rangle \). Notice that
there is now a second quartic term with co-efficient \( v \). If \( v > 0 \), configurations with \( \Phi_{\alpha}^2 = 0 \) are preferred: in such configurations \( \Phi_{\alpha} = n_{1\alpha} + in_{2\alpha} \), where \( n_{1,2\alpha} \) are two equal-length orthogonal vectors. Then from (13) and (6) it is
easy to see that the physical spins possess spiral order in the magnetically
ordered state in which \( \Phi_{\alpha} \) is condensed. A spiral state is illustrated in Fig ??, and we will have more to say about this state in Section ???. For the case
\( v < 0 \), the optimum configuration has \( \Phi_{\alpha} = n_{\alpha}e^{i\theta} \) where \( n_{\alpha} \) is a real vector: this leads to a magnetically ordered state with spins polarized collinearly in a
spin density wave at the wavevector \( Q \).

1.3 Quantum criticality

We will restrict our discussion here to the critical point described by \( S_{\phi} \).
Similar results apply to \( S_{\Phi} \) for the parameter regime in which it exhibits a
second order transition [31]. Experimentally, the results below are relevant
to materials that can be tuned across the magnetic ordering transition by
applied pressure (such as TlCuCl₃ [8]), or to materials which happen to be
near a critical point at ambient pressure (such as LaCuO₂₅ [32]).

The field theory \( S_{\phi} \) is actually a familiar and well-studied model in the
context of classical critical phenomena. Upon interpreting \( \tau \) as a third spatial
co-ordinate, \( S_{\phi} \) becomes the theory of a classical O(3)-invariant Heisenberg
ferromagnet at finite temperatures (in general a \( d \) dimensional quantum anti-
ferromagnet will map to a \( d + 1 \) dimensional classical Heisenberg ferromagnet
at finite temperature [33]). The Curie transition of the Heisenberg ferromag-
net then maps onto the quantum critical point between the paramagnetic
and Néel states described above. A number of important implications for the
quantum problem can now be drawn immediately.

The theory \( S_{\phi} \) has a ‘relativistic’ invariance, and consequently the dynamic
critical exponent must be \( z = 1 \). The spin correlation length will diverge at the
quantum critical point with the exponent \( \nu = 0.7048(30) \). The spin gap
of the paramagnet, $\Delta$, vanishes as $\Delta \sim (\lambda - \lambda_c)^{-\nu}$, and this prediction is in excellent agreement with the numerical study of the dimerized antiferromagnet [28].

A somewhat more non-trivial consequence of this mapping is in the structure of the spectrum at the critical point $\lambda = \lambda_c$. At the Curie transition of the classical ferromagnet it is known [35] that spin correlations decay as $\sim 1/p^{2-\eta}$, where $p$ is the 3-component momentum in the 3-dimensional classical space. We can now analytically continue this expression from its $p_z$ dependence in the third classical dimension to the real frequency, $\omega$, describing the quantum antiferromagnet. This yields the following fundamental result for the dynamic spin susceptibility, $\chi(k, \omega)$, at the $T = 0$ quantum critical point of the coupled-dimer antiferromagnet:

$$\chi(k, \omega) \sim \frac{1}{(c_x^2 k_x^2 + c_y^2 k_y^2 - (\omega + i\epsilon)^2)^{1-\eta/2}},$$

where $\epsilon$ is a positive infinitesimal. Note that in (15) the momentum $k$ is measured from the $(\pi, \pi)$ ordering wavevector of the Néel state. The exponent $\eta$ is the same as that of the classical Heisenberg ferromagnet, and has a rather small value [34]: $\eta \approx 0.03$. However, the non-zero $\eta$ does make a significant difference to the physical interpretation of the excitations at the critical point. In particular note that $\text{Im} \chi(k, \omega)$ does not have a pole at any $k$, but rather a continuum spectral weight above a threshold energy [36, 37]

$$\text{Im} \chi(k, \omega) \sim \text{sgn}(\omega) \sin \left( \frac{\pi \eta}{2} \right) \frac{\theta \left( |\omega| - \sqrt{c_x^2 k_x^2 + c_y^2 k_y^2} \right)}{(\omega^2 - c_x^2 k_x^2 - c_y^2 k_y^2)^{1-\eta/2}},$$

where $\theta$ is the unit step function. This indicates there are no quasiparticles at the critical point, and only a dissipative critical continuum.

There is also some very interesting structure in the quantum critical dynamic response at nonzero $T$ [36,37]. Here, one way to understand the physics is to approach the critical point from the paramagnetic side ($\lambda < \lambda_c$). As we noted earlier, the paramagnetic phase has well-defined ‘triplon’ or ‘spin exciton’ excitations $t_\alpha$, and these have an infinite lifetime at $T = 0$. At $T > 0$, thermally excited $t_\alpha$ quasiparticles will collide with each other via their scattering amplitude, $u$, and this will lead to a finite lifetime [37, 38]. Now approach $\lambda = \lambda_c$. The renormalization group analysis of $S_\phi$ tells us that the quartic coupling $u$ approaches a fixed point value in the critical region. This means that $u$ is no longer an arbitrary parameter, and an appropriately defined $t_\alpha$ scattering amplitude must also acquire universal behavior. In particular, the $t_\alpha$ lifetime is determined by the only energy scale available, which is $k_B T$. So we have the remarkable result that the characteristic spin relaxation time is a universal number times $\hbar/(k_B T)$. More precisely, we can write for the local dynamic spin susceptibility $\chi_L(\omega) = \int d^2k \chi(k, \omega)$ the universal scaling form
\[ \text{Im} \chi_L(\omega) = T^n F \left( \frac{\hbar \omega}{k_B T} \right) \]  

(17)

Here \( F \) is a universal function which has the limiting behaviors

\[ F(\omega) \sim \begin{cases} \omega, & |\omega| \ll 1 \\ \text{sgn}(\omega)|\omega|, & |\omega| \gg 1 \end{cases} \]  

(18)

Note that \( F \) has a smooth linear behavior in the regime \( |\hbar \omega| \ll k_B T \), and this is similar to any simple dissipative system. The difference here is that the coefficient of dissipation is determined by \( k_B T \) alone.

The quantum critical behavior described here is expected to apply more generally to other correlated electron systems, provided the critical theory has non-linear couplings which approach fixed point values.

\section*{2 Influence of an applied magnetic field}

An important perturbation that can be easily applied to antiferromagnets in the class discussed in Section 1 is a uniform magnetic field. The Zeeman energy in available fields can often be comparable to the typical antiferromagnetic exchange constant \( J \), and so the ground state can be perturbed significantly. It is therefore of interest to understand the evolution of the phase diagram in Fig 4 under an applied field of arbitrary strength.

We are interested here in the evolution of the ground state as a function of \( B \) where the Hamiltonian \( H_d \) in (1) is transformed as

\[ H_d \rightarrow H_d - \sum_j B \cdot S_j. \]  

(19)

Most of the basic features can actually be understood quite easily in a simple extension of the self-consistent Hartree-Fock theory of bond bosons that was discussed in Section 1.2. Under the transformation (19), it is easily seen from (6) that

\[ H_d \rightarrow H_d + iB_\alpha \sum_{\ell \in A} \epsilon_{\alpha\beta\gamma} t^\dagger_{\ell\beta} t_{\ell\gamma} \]  

(20)

The presence of a non-zero \( B \) breaks spin rotation invariance and so all the self-consistent expectation values of operator bilinears have to reflect this reduced symmetry in the Hartree-Fock theory. Apart from this the mechanics of the computation mostly remain the same. However, for stronger fields, it is sometimes necessary to allow for broken translational symmetry in the expectation values, as the ground state can acquire a modulated structure.

We will discuss the results of such an analysis in weak and strong fields in the following subsections.
Fig. 5. Evolution of the phases of Fig 4 under a weak field $B$ (magnetization plateau at large $B$, appearing in Fig 5, are not shown). The paramagnetic phase has exactly the same ground state wavefunction as that at $B = 0$. The phase boundary behaves like $B \sim (\lambda_c - \lambda)^c$. The $B$ field is oriented vertically upwards, and the static moments in the canted phase can rotate uniformly about the vertical axis. The phase boundary at non-zero $B$ is described by the $z = 2$ dilute Bose gas quantum critical theory. The phase diagram of TlCuCl$_3$ in applied pressure and magnetic field looks similar to the one above [10]. The corresponding phase diagram of the field-induced magnetic ordering transition of a superconductor (rather than a Mott insulator) has been investigated recently [39], and successfully applied to experiments on the doped cuprates; this phase diagram of the superconductor has significant differences from the one above.

2.1 Weak fields

For weak fields applied to the paramagnet (specifically, for fields $B < \Delta$, the influence of (20) can be understood exactly. The coupling to $B$ involves an operator which commutes with the remaining Hamiltonian (the total spin), and hence the wavefunction of the ground state remains insensitive to the value of $B$. The same applies to the wavefunctions of the excited states. However, the excited states can have non-zero total spin and so their energies do depend upon $B$. In particular the triplet $t_\alpha$ quasiparticle with energy (3) or (10) carries total spin $S = 1$, and consequently we conclude that this triplet splits according to

$$\varepsilon_k \rightarrow \varepsilon_k - mB$$

with $m = 0, \pm 1$. Note that the lowest energy quasiparticle (with $m = 1$) has a positive energy as long as $B < \Delta$, and this is required for the stability of the paramagnet. So the phase boundary of the paramagnetic phase is exactly $B = \Delta$, and using $\Delta \sim (\lambda_c - \lambda)^{2\nu}$, we can sketch the boundary of the paramagnetic phase as in Fig 5.

What happens beyond the paramagnetic phase? As in Section 1.2, we answer this question by using the transformation (11), and by examining the analog of $S_\phi$ under a non-zero $B$. Using (20), and integrating out $\pi_\alpha$, we now find that the action $S_\phi$ in (12) remains unchanged apart from the mapping
The action (12), (22) can now be analyzed by a traditional small fluctuation analysis about $\phi_\alpha = 0$. Let us assume that $B = (0, 0, B)$ is oriented along the $z$ axis. Then the coefficient of $\phi_z^2$ is $s$, while that of $\phi_x^2 + \phi_y^2$ is $s - B^2$. This suggests that we focus only on the components of $\phi_\alpha$ in the plane orthogonal to $B$, and integrate out the component of $\phi_\alpha$ along the direction of $B$. Indeed, if we define

$$\Psi = \frac{\phi_x + i\phi_y}{\sqrt{B}}$$

and integrate out $\phi_z$, then we obtain from (12), (22) the effective action for $\Psi$:

$$S_\Psi = \int d^2r d\tau \left[ \Psi^* \partial_\tau \Psi + \frac{\mu^2}{2B} |\partial_\tau \Psi|^2 + \frac{\mu^2}{2B} |\partial_\tau \Psi|^2 - \mu |\Psi|^2 \right].$$

Here, $\mu = (s - B^2)/2B$, and we have retained only leading order temporal and spatial gradients and the leading dependence of $u$. Clearly, this is the theory of a Bose gas in the grand canonical ensemble at a chemical potential $\mu$, with a repulsive short-range interaction. At $T = 0$, and $\mu < 0$, such a theory has a ground state which is simply the vacuum with no Bose particles. Here, this vacuum state corresponds to the spin gap antiferromagnet, and the $B$-independence of the ground state of the antiferromagnet corresponds here to the $\mu$ independence of the ground state of $S_\Psi$. There is an onset of a finite density of bosons in $S_\Psi$ for $\mu > 0$, and this onset therefore corresponds to the quantum phase transition in the antiferromagnet at $B = \Delta$. So we must have $\mu = 0$ in $S_\Psi$ at precisely the point where $B = \Delta$: the value of $\mu$ quoted above shows that this is true at zeroth order in $u$, and higher order terms in $u$ must conspire to maintain this result.

The above analysis makes it clear that the $\mu \geq 0$ region of $S_\Psi$ will describe the quantum phase transition out of the paramagnet at non-zero $B$. This transition is merely the formation of a Bose-Einstein condensate of the $m = 1$ component of the triplon bosons. For $\mu > 0$ we have a finite density of $\Psi$ bosons which Bose condense in the ground state, so that $\langle \Psi \rangle \neq 0$. From (23) we see that this Bose condensation corresponds to antiferromagnetic order in the plane perpendicular to $B$. Indeed, the phase of this Bose condensate is simply the orientation of the spins in the $x,y$ plane, and so here this phase is directly observable. Further, by taking derivatives of (19) and $S_\Psi$ w.r.t. $B$, we see that the density of bosons is proportional to the magnetization per spin, $\Omega$, in the direction parallel to $B$:

$$\Omega = \frac{1}{N} \sum_j \langle S_{jz} \rangle \propto \langle |\Psi|^2 \rangle,$$

where $N$ is the total number of spins. Consequently, the average magnetic moments in the non-paramagnetic phase are in a ‘canted’ configuration, as
shown in Fig 3. The quantum phase transition between the paramagnet and the canted state is described by the theory of the density onset in a Bose gas: this theory has \( z = 2, \nu = 1/2, \) and an upper critical dimension of \( d = 2 \) \[42\] \[41\].

We conclude this section by noting that interesting recent work \[43\] has examined the Bose-Einstein condensation of the \( m = 1 \) triplon bosons in a random potential. This is achieved by studying \( \text{Th}_{1-x}K_x\text{CuCl}_3 \), where the stoichiometric disorder among the non-magnetic ions acts as a random potential on the triplons.

### 2.2 Strong fields

We have seen above that applying a magnetic field eventually leads to the onset of a ferromagnetic moment in the directions of the applied field. How does this moment evolve as we continue to increase the field? Eventually, \( B \) will become so large that it pays to have all the spins polarized in the direction of the field: this corresponds to a saturation in the magnetization, and making \( B \) even stronger will not change the ground state. In terms of the \( t \) bosons, this fully polarized state, \(|FP\rangle\), with \( \Omega = 1/2 \), is seen from (20) or (4) to correspond exactly to

\[
|FP\rangle = \prod_\ell \left( t_{\ell x}^\dagger + i t_{\ell y}^\dagger \right) \frac{1}{\sqrt{2}} |0\rangle.
\]

So there must be at least one more quantum phase transition as \( B \) is increased: this is transition from the \(|FP\rangle\) state at very large \( B \) to a state with a continuously varying ferromagnetic moment which eventually reaches the saturation value from below.

A theory for the transition away from the \(|FP\rangle\) state with decreasing \( B \) can be developed using methods very similar to those used in Section 1.2 and 2.1. We treat the quartic terms in (7) in a Hartree-Fock approximation, and examine small fluctuations away from the \(|FP\rangle\) state. These are dominated by excitation which create \( t_z \) quanta (which have \( m = 0 \)) on the dimers, and so the effective theory is expressed in terms of

\[
\bar{\Psi}^\dagger \sim t_z^\dagger (t_x - i t_y).
\]

Indeed, it is not difficult to see that the resulting theory for \( \bar{\Psi} \) has exactly the same form as (24). Now the \( \mu \) for \( \bar{\Psi} \) decreases with increasing \( B \), and we have \( \mu = 0 \) at the critical field at which \(|FP\rangle\) first becomes the ground state. Furthermore, \( \langle\bar{\Psi}^2\rangle \) now measures the deviation away from \( \Omega = 1/2 \). Apart from this ‘inversion’ in the field axis, it is clear that the universality class of the present transition is identical to that discussed in Section 2.1.

A further possibility for a plateau in the value of \( \Omega \) with increasing \( B \) is worth mentioning \[44\], as analogs are realized in \( \text{SrCu}_2(\text{BO}_3)_2 \) \[45\] and
NH$_4$CuCl$_3$ [40]. So far we have found plateaus at $\Omega = 0$ for $B < \Delta$, and at $\Omega = 1/2$ for large $B$. For the $\Omega = 1/2$ state we had every dimer with a $(t_1^a + it_1^b)/\sqrt{2}$ boson. Now imagine that these bosons form a Wigner-crystalline state so that there are $p$ such bosons for every $q$ dimers; here $0 \leq p \leq q$, $q \geq 1$, are integers. Such a state will have $\Omega = p/(2q)$, and breaks the translational symmetry of the underlying dimer antiferromagnet such that there are $q$ dimers per unit cell (or $2q$ spins per unit cell). The energy gap towards boson motion in the Wigner crystal (i.e. its incompressibility) will ensure that $\Omega$ is stable under small variations of $B$. In this manner we can obtain a magnetization plateau at $\Omega = p/(2q)$ in a state with a unit cell of $q$ dimers.

We summarize the considerations of this subsection in Fig 6, showing a possible evolution of $\Omega$ in a model similar to $H_d$ in [1]. As we have already noted, the plateau onset transitions at $\Omega = 0$ and $\Omega = 1/2$ are both described by the $z = 2$ dilute Bose gas theory [24]. The transitions in and out of other fractional plateaus are potentially more complicated because these involve spontaneous breaking of translational symmetry. The translation symmetry could be restored at the same point at which there is onset of superfluid order—this is possibly a first order transition with a jump in the value of $\Omega$. Alternatively, there could be an intermediate ‘supersolid’ phase, in which case the plateau transition has the same broken translational symmetry on both sides of it, placing it also in the class of [24].
References

13. Insulators with an even number of electrons per unit cell can be adiabatically connected to band insulators, and so some readers may object to calling such materials ‘Mott insulators’. However, the very different energy scales of the spin and charge excitations in the experimental systems are best understood in the framework of the Mott theory, and one regards the dimerization as a small, low energy, deformation. Following widely accepted practice, we will continue to label these materials Mott insulators.
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