# Bose-Einstein condensation in magnetic insulators

The Bose–Einstein condensate (BEC) is a fascinating state of matter predicted to occur for particles obeying Bose statistics. Although the BEC has been observed with bosonic atoms in liquid helium and cold gases, the concept is much more general. We here review analogous states, where excitations in magnetic insulators form the BEC. In antiferromagnets, elementary excitations are magnons, quasiparticles with integer spin and Bose statistics. In certain experiments their density can be controlled by an applied magnetic field leading to the formation of a BEC. Furthermore, interactions between the excitations and the interplay with the crystalline lattice produce very rich physics compared with the canonical BEC. Studies of magnon condensation in a growing number of magnetic materials thus provide a unique window into an exciting world of quantum phase transitions and exotic quantum states, with striking parallels to phenomena studied in ultracold atomic gases in optical lattices.

# THIERRY GIAMARCHI<sup>1\*</sup>, CHRISTIAN RÜEGG<sup>2\*</sup> AND OLEG TCHERNYSHYOV<sup>3\*</sup>

<sup>1</sup>DPMC-MaNEP, University of Geneva, 24 Quai Ernest Ansermet, 1211 Geneva 4, Switzerland

<sup>2</sup>London Centre for Nanotechnology and Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK

<sup>3</sup>Department of Physics and Astronomy, Johns Hopkins University, Baltimore, Maryland 21218, USA

\*e-mail: thierry.giamarchi@physics.unige.ch; c.ruegg@ucl.ac.uk; olegt@jhu.edu

Not long after Bose and Einstein described quantum statistics of photons<sup>1</sup> and atoms<sup>2</sup>, Bloch applied the same concept to excitations in solids. He treated misaligned spins in a ferromagnet as magnons, particles with integer spin and bosonic statistics, to describe the reduction of spontaneous magnetization by thermal fluctuations<sup>3</sup>. Matsubara and Matsuda pointed out an exact correspondence between a quantum antiferromagnet and a lattice Bose gas<sup>4</sup>. It is thus natural to ask whether these bosons can undergo Bose-Einstein condensation and become superfluid. The answer is yes. The question of order in spin systems and the possibility of Bose-Einstein condensation of magnons has been investigated theoretically for a number of quantum antiferromagnets<sup>5-13</sup>. Although the concept of a BEC can be applied in simple spin systems, in practice factors such as the large values of exchange constants and the presence of anisotropies violating rotational symmetry may restrict their usefulness. However, the analogy between spins and bosons has proven to be very fruitful in those antiferromagnets where closely spaced pairs of spins S = 1/2 form dimers with a spin-singlet (S = 0) ground state and triplet (S = 1)bosonic excitations called triplons. The triplons are similar to magnetic excitations of an ordered antiferromagnet, the magnons;

in particular, they have the same quantum numbers, so the two terms are sometimes used interchangeably. In the dimer systems, Bose–Einstein condensation has been predicted to occur<sup>7</sup> and was experimentally observed<sup>8,14</sup> in the magnetic insulator TlCuCl<sub>3</sub>. This started a flurry of activity on the subject and the hunt for such transitions in other magnetic materials.

Other examples of condensates from elementary excitations were recently found in magnetic thin films<sup>15</sup>, semiconductor microcavities<sup>16</sup>, <sup>3</sup>He (refs 17,18) and models for ferromagnets<sup>19,20</sup>. Lattice bosons and several transitions occurring in these systems have been studied extensively in ultracold atomic gases in optical lattices<sup>21–24</sup>. Here we present an overview of recent developments in the field and discuss how quantum antiferromagnets offer several advantages that distinguish them from other model systems in which BEC occurs. We also discuss ways to go beyond the physics of simple Bose–Einstein condensation and to look at the fascinating new quantum phases of interacting bosons on a lattice.

# **BOSONS IN MAGNETS**

Let us illustrate the basics of a magnon BEC in real dimerized antiferromagnets, such as the extensively studied TlCuCl<sub>3</sub> (refs 8,11,12,14,25–33) and BaCuSi<sub>2</sub>O<sub>6</sub> (refs 34–40). The lattice of magnetic ions in such materials (Fig. 1a) can be visualized as a set of dimers, pairs of copper ions Cu<sup>2+</sup> carrying S = 1/2 each and interacting via Heisenberg exchange:

$$\mathcal{H} = \sum_{i} J_0 \mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i} + \sum_{\langle mnij \rangle} J_{mnij} \mathbf{S}_{m,i} \cdot \mathbf{S}_{n,j} - g \mu_B H \sum_{\langle ni \rangle} S_{m,i}^z, \qquad (1)$$

where  $\mu_B$  is the Bohr magneton, H denotes an external magnetic field in the z-direction and i, j number dimers and m, n = 1, 2 their magnetic sites. The intradimer exchange is the strongest

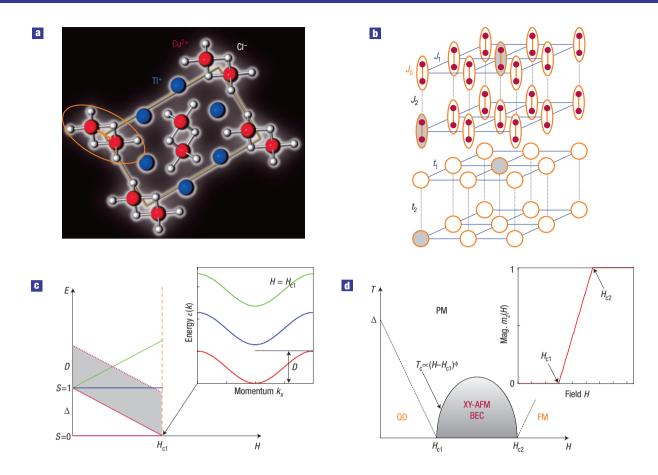


Figure 1 BEC of magnons in dimerized quantum antiferromagnets. **a**, Dimers in the real material TICuCl<sub>3</sub> with S=1/2 from Cu<sup>2+</sup> ions and superexchange via Cl<sup>-</sup> (refs 8,11,12,14,25,26,99). **b**, Dimers on a square lattice with dominant antiferromagnetic intradimer interaction  $J_0$  and interdimer interactions  $J_i$ . Triplet states (grey, top) are mapped onto quasiparticle bosons (triplons, bottom). **c**, Zeeman splitting of the triplet modes with gap  $\Delta$  and bandwidth D at  $\mathbf{k}_0 = (\pi/a, \pi/a)$ . Inset, Dispersion of triplons at the critical field  $H_{c1}$  (refs 11,12,14,25,26). **d**, Resulting phase diagram with paramagnetic (PM), quantum disordered (QD) and field-aligned ferromagnetic (FM) phases and canted-antiferromagnetic (XY-AFM) phase, where a magnon BEC occurs. Close to  $H_{c1}$  and  $H_{c2}$  the phase boundary follows a power law  $T_c \propto (H - H_{c1})^{\phi}$  with a universal exponent  $\phi = 2/3$  for a magnon BEC<sup>7,8</sup>. Inset, Magnetization curve  $m_z(H)$  for a three-dimensional dimer spin system with a plateau at  $m_z = 0$ , 1 (gapped)<sup>8,36,37</sup>.

interaction, which is antiferromagnetic,  $J_0 > 0$ , so that an isolated dimer has a ground state with total spin S = 0 and a triply degenerate excited state of energy  $J_0$  and spin S = 1 (Fig. 1c). It is convenient to identify the triplet state with the presence of a triplon, a bosonic particle with S = 1, and the singlet state with the absence of a triplon; see Fig. 1b. As long as interdimer interactions are weak, the ground state consists of non-magnetic singlets. It remains disordered down to absolute zero temperature without long-range magnetic order. The triplon excitations are made mobile by weak interdimer couplings  $J_{1,2,...}$ , from a sum over single-ion interactions  $J_{mnij}$ ; see Fig. 1b. For dimers forming a square lattice (for simplicity), the energy of a triplon with spin projection  $S^z = 0, \pm 1$  to first order in  $J_1$  is

$$\varepsilon(\mathbf{k}) = J_0 + J_1[\cos(k_x a) + \cos(k_y a)] - g\mu_B H S^z, \tag{2}$$

where  $\mathbf{k} = (k_x, k_y)$  is the quasiparticle wavevector, a is the lattice constant and  $D = 4J_1$  is the bandwidth; see Fig. 1c. The energy–momentum dependence (dispersion) of the triplons and singlet–triplet correlations have been measured directly by inelastic neutron scattering<sup>25,34,35,41</sup>.

The Zeeman term  $-g\mu_{\rm B}HS^z$  controls the density of triplons. As the magnetic field increases, the excitation energy of triplons

with  $S^z=+1$  is lowered and eventually crosses zero, as shown in Figs 1c and 2a. This defines two critical magnetic fields  $H_{\rm cl}$  and  $H_{\rm c2}$  in the phase diagram; see Fig. 1d. At zero temperature, below  $H_{\rm cl}$  the magnetization  $m_z(H)$  is zero and only singlets exist. Between  $H_{\rm cl}$  and  $H_{\rm c2}$  the magnetization increases with increasing field as more triplons appear in the ground state owing to the increased gain in Zeeman energy; see Fig. 1c. Above  $H_{\rm c2}$  each site is occupied by a triplon and the magnetization saturates at one per dimer.

The bosonic nature of triplons is guaranteed by the simple fact that spin operators of two different dimers commute. However, because a dimer can hold at most one triplon, the bosonic picture requires the introduction of a hard-core constraint to exclude states with more than one quasiparticle per dimer. The constraint, which can be interpreted as a strong short-range repulsion between the bosons, poses a difficult theoretical problem. However, close to  $H_{\rm cl}$  their density is small and collisions between bosons are rare; in this limit interactions can be fully taken into account. By particle—hole symmetry, a similar simplification occurs near  $H_{\rm c2}$ .

# **BEC OF TRIPLONS**

The nature of the ground state above  $H_{c1}$ , its interpretation as a BEC of magnetic quasiparticles and the tuning of the particle density

become more accessible if the spin hamiltonian in equation (1) is rewritten in the second-quantized form<sup>7,8</sup>

$$\mathcal{H} = \sum_{i} (J_0 - h) a_i^{\dagger} a_i + \sum_{i,j} t_{ij} a_i^{\dagger} a_j + \frac{1}{2} \sum_{i,j} U_{ij} a_i^{\dagger} a_j^{\dagger} a_j a_i \qquad (3)$$

where  $a_i^{\dagger}$  ( $a_i$ ) creates (annihilates) a boson on dimer i and  $h = g \mu_{\rm B} H$  is the field in energy units. The term  $t_{ij}$ , generated by the coupling of the transverse spin components  $S_{m,i}^x S_{n,j}^x + S_{m,i}^y S_{n,j}^y$ , describes hopping between sites i and j, thus endowing the triplons with kinetic energy; see Fig. 1b.  $U_{ij}$  is the repulsion energy arising from the longitudinal term  $S_{m,i}^z S_{n,j}^z$  when two triplons occupy neighbouring dimers i and j. The O(2) symmetry of the spin hamiltonian (1) translates into a U(1) symmetry of the boson hamiltonian (3) and the density of triplons is directly controlled by the applied magnetic field, which acts as a chemical potential. (We refer those purists who object to the interpretation of the magnetic field as a chemical potential to Shakespeare's *Romeo and Juliet* II.2.43: "What's in a name? That which we call a rose By any other word would smell as sweet".)

Below  $H_{c1}$  the ground state is a quantum-disordered paramagnet formed by the singlet sea (triplon vacuum) and can be approximated by the direct product of singlet states on each dimer,  $|\psi\rangle_i = |S, S^z\rangle_i$  with  $S = S^z = 0$ . Once the spin gap is closed at  $H_{c1}$ , a Bose condensate is formed. Because the bottom of the triplon band is located at a non-zero wavevector  $\mathbf{k}_0 = (\pi/a, \pi/a)$  (Fig. 1c), the wavefunction of the condensate varies in space as  $\exp(i\mathbf{k}_0 \cdot \mathbf{r})$ . In this phase the state of an individual dimer at position  $\mathbf{r}$  is well approximated by a coherent superposition of the singlet and the  $S^z = +1$  triplet:  $|\psi\rangle_i = \alpha_i(H)|0,0\rangle_i + \beta_i(H)|1,1\rangle_i$ , where the amplitudes  $\alpha_i$  and  $\beta_i$  depend on the magnetic field H (refs 7,11,12).

In spin language, the condensate corresponds to magnetic order, formed by the transverse components  $\langle S_i^x \rangle$  and  $\langle S_i^y \rangle$ , which spontaneously breaks the rotational O(2) symmetry of the spin hamiltonian (1). To make the analogy with the traditional BEC manifest, we can form a U(1) order parameter  $\langle S_i^x + iS_i^y \rangle$ . The phase corresponding to the angle of the spin in the XY plane is thus the phase of the wavefunction in boson language. At  $H_{c1}$ the paramagnetic phase at low fields makes a transition into a canted antiferromagnet with long-range magnetic order in the plane perpendicular to the field<sup>7</sup> (Figs 1d, 3a). The staggering of the transverse components of magnetization reflects a non-zero wavevector  $\mathbf{k}_0$  of the condensate. The critical properties of the magnet in the vicinity of this phase transition are governed by the quantum critical point (QCP) of the BEC universality class located at T = 0 and  $H = H_{c1}$ . Indeed, close to  $H_{c1}$  the bosons are extremely diluted, weakening the effects of the hard-core interactions. Close to the QCP, the phase boundary  $T_c(H)$  follows<sup>7</sup> a power law  $T_c \propto (H - H_{c1})^{\phi}$  with a universal critical exponent  $\phi = z/d$ , which depends only on the dimensionality d and dynamical critical exponent z = 2 for a quadratic triplon energy band. The upper critical dimension for the QCP is  $d_c = 2$ .

The BEC quantum phase transition has been observed in a growing number of dimer-based magnetic insulators, such as  $A\text{CuCl}_3$  (A = Tl, K, NH<sub>4</sub>), BaCuSi<sub>2</sub>O<sub>6</sub>, Cu(NO<sub>3</sub>)<sub>2</sub>·2·5D<sub>2</sub>O, Cs<sub>3</sub>Cr<sub>2</sub>Br<sub>9</sub>, (CH<sub>3</sub>)<sub>2</sub>CHNH<sub>3</sub>CuCl<sub>3</sub> and (C<sub>4</sub>H<sub>12</sub>N<sub>2</sub>)Cu<sub>2</sub>Cl<sub>6</sub> (refs 8,14, 36,37,40,42–49). The physics of triplon condensation, presented above for a weakly coupled dimer system, remains essentially unchanged, because these QCPs are in the same universality class. Quasi-one-dimensional arrangements of S = 1 moments in spin chains, for example Haldane chains, and even field-saturated frustrated antiferromagnets can be described within the same framework, as has been successfully done for nickel-based materials and Cs<sub>2</sub>CuCl<sub>4</sub>, respectively<sup>50–54</sup>.

Table 1 Correspondence between a Bose gas and a quantum antiferromagnet.

| Bose gas   | Antiferromagnet  |
|--|--|
| Particles  | Spin excitations ( $S=1$ quasiparticles)                   |
| Boson number N   | Spin component S <sup>z</sup>                              |
| Charge conservation <i>U</i> (1)                           | Rotational invariance O(2)                                 |
| Condensate wavefunction $\langle \psi(\mathbf{r}) \rangle$ | Transverse magnetic order $\langle S_i^x + iS_i^y \rangle$ |
| Chemical potential $\mu$                                   | Magnetic field H   |
| Superfluid density $\rho_s$                                | Transverse spin stiffness                                  |
| Mott insulating state                                      | Magnetization plateau                                      |

Experimentally, the static and dynamic magnetic properties have been studied at and around the BEC QCPs in such materials by many experimental techniques. The evolution of magnetic excitations across the QCP at  $H_{c1}$  in a three-dimensional network of dimers was studied in TlCuCl<sub>3</sub>; see Figs 1c, 2a,c<sup>11,12,14,26</sup>. The softening of the triplet mode  $S^z = +1$  in the quantum disordered phase, Fig. 2a, is followed by a dramatic change in the nature of the excitation spectrum above the critical field  $H_{c1}$ . In particular, as can be expected in a system with a spontaneously broken XY symmetry in the plane perpendicular to the applied magnetic field, a Goldstone mode appears with a soundlike dispersion  $\hbar \omega(\mathbf{k}) \sim \hbar s |\mathbf{k} - \mathbf{k}_0|$ , where s is a velocity (refs 11,12).

Historically, the temperature dependence of the magnetization  $m_z(T)$  at fixed field  $H > H_{c1}$  in TlCuCl<sub>3</sub> (Fig. 2d) and the scaling of the critical temperature  $T_c(H)$  were the key experimental observations that confirmed the theoretical model<sup>7,8</sup> of the magnon BEC in dimerized quantum antiferromagnets. The minimum and cusp in  $m_z(T)$  at the finite-temperature transition cannot be explained within simple mean-field theory<sup>55</sup>, but are a consequence of magnon condensation<sup>7,8</sup> (and as such also occur in lowerdimensional magnets). Even in the one-dimensional case, where the condensate is disordered by thermal and quantum fluctuations, similar minima in the magnetization can occur, but now as a simple crossover<sup>56</sup>. Comprehensive data from bulk measurements establish the phase diagram and critical exponents for BaCuSi<sub>2</sub>O<sub>6</sub> (Fig. 2b). The temperature crossover exponent is found to be  $\phi = 2/3$ , as expected for a BEC QCP in three dimensions. However, below 1 K and down to 35 mK the phase boundary unexpectedly becomes linear, indicating<sup>37–40</sup> that  $\phi = 1$ . We discuss this anomaly below.

## COMPARISON WITH OTHER BOSON SYSTEMS

The correspondence between a Bose gas and an antiferromagnet is summarized in Table 1. Despite many similarities, there are also a few important differences between triplons in a magnet and atoms in a Bose gas<sup>21–24</sup>. Most importantly, the number of atoms is usually controlled directly (microcanonical ensemble), whereas the number of triplons is typically set by the magnetic field acting like a chemical potential (canonical ensemble).

Considerable differences also exist at the practical level. Triplons are much lighter and have a much higher density than atomic gases. As a result, condensates survive to much higher temperatures: kelvins in magnets, even room temperature in some cases<sup>15</sup>, as opposed to nanokelvins in atomic gases. As solid-state systems, they also allow for a variety of static and dynamic probes (magnetization, specific heat, NMR, neutrons and so on). On the other hand, the cold atomic systems enable us potentially to study some out-of-equilibrium situations that are hard to maintain in a solid-state device because the condensate is still strongly coupled to the dissipative environment.

From the point of view of realization, systems with cold atoms have a high degree of control and tunability in terms of the interactions and structure. Indeed, the structure of the lattice and

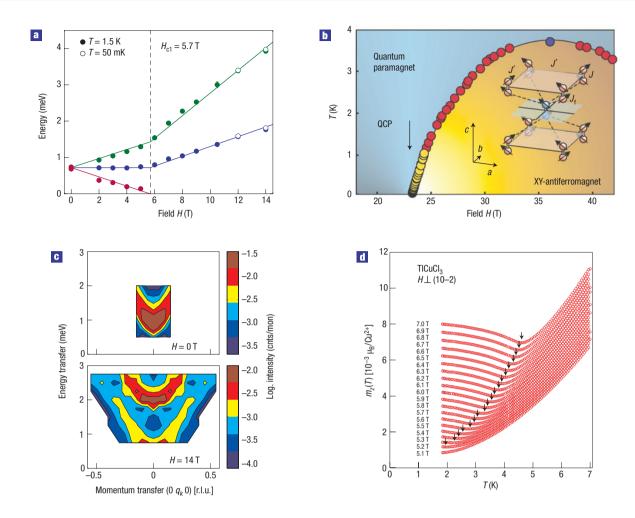


Figure 2 Experimental results on the magnon BEC. a, Zeeman splitting of the triplet modes in TICuCl<sub>3</sub> up to  $H > H_{c1}$  measured by inelastic neutron scattering<sup>14,31,32</sup>. b, Phase diagram of BaCuSi<sub>2</sub>O<sub>6</sub> measured by torque magnetization, magneto-caloric effect and specific heat<sup>36,37</sup>. Dimensional reduction was reported in this material, with a crossover from the three-dimensional BEC critical exponent  $\phi = 2/3$  to  $\phi = 1$  for two dimensions at temperatures close to the QCP<sup>37</sup>. c, Excitations in the BEC of triplons realized in TICuCl<sub>3</sub> (refs 14,31,32). Goldstone mode with linear dispersion around  $\mathbf{k}_0$ , corresponding to  $q_k = 0$  in the notation of ref. 14. Spin anisotropy generally leads to a spin gap in real materials<sup>31–33</sup>. d, Temperature dependence of the magnetization  $m_z(T)$  in TICuCl<sub>3</sub> for fixed magnetic field H, as indicated<sup>8</sup>. Minima at the finite-temperature phase transition (vertical arrows), as expected for a BEC of triplons.

the hopping of the bosons can directly be controlled by varying the strength of an optical lattice. The interaction can also be controlled via a Feshbach resonance, enabling us in principle to design our pet hamiltonian. In spin systems the parameters can be changed only in a limited way by the application of pressure or by changing the chemical composition. Efforts in quantum chemistry have produced good realizations of three-, two- and one-dimensional dimer materials, for example TlCuCl<sub>3</sub>, BaCuSi<sub>2</sub>O<sub>6</sub> and (C<sub>5</sub>H<sub>12</sub>N)<sub>2</sub>CuBr<sub>4</sub>, respectively. In addition, by the very nature of the mapping from spins to bosons, spin systems offer a definite advantage in reaching the limit of strong on-site repulsion as well as the effects of interactions between nearest neighbours (see the next section). Spin systems are therefore an optimal starting point to study situations for which these ingredients are important.

The cold atomic systems have the important advantage that the phase U(1) symmetry is exact. In the magnets, the corresponding O(2) symmetry in the plane perpendicular to the magnetic field can be broken by weak anisotropic interactions (crystalline anisotropies, spin–orbit coupling and dipolar interactions). As a matter of fact, they may always be present in real magnetic

materials at some, preferably low, energy scale (compared with interactions  $J_{mnij}$ )<sup>31,57,58</sup>, or it may be challenging experimentally to apply the magnetic field exactly along a symmetry direction. Even when symmetry-breaking terms are weak, they become important at low temperatures and modify the physics in the vicinity of the QCP. Although such additional terms in the spin hamiltonian should be part of any detailed experimental characterization, many of the currently studied magnetic materials provide a clean experimental window where critical exponents, effects of dimensionality and so on can be investigated in the anticipated physical limits <sup>8,37,38,57</sup>. The physics of BEC may also be altered in the presence of coupling between spins and lattice distortions. Fortunately, a thorough understanding of BEC in O(2)-symmetric magnets enables us to treat theoretically small deviations from the idealized case <sup>30,32,33,59-62</sup>.

Finally, these spin systems provide an excellent opportunity to study the critical behaviour in the immediate vicinity of a QCP because the high degree of homogeneity in boson density offered by crystalline solids is difficult to attain in the presence of a trap potential confining an atomic gas.

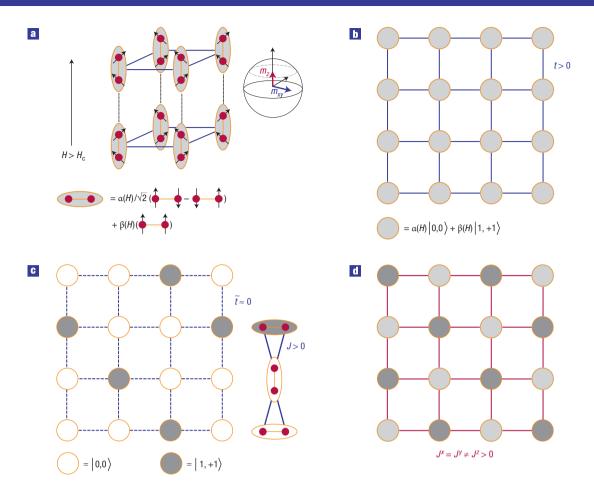


Figure 3 Lattice boson phases. **a**, Canted antiferromagnetic state at  $H_{c1} < H < H_{c2}$  in a BEC of magnons. Longitudinal  $m_z$  (see Fig. 1d) and transverse (*XY*) magnetization  $m_{xy}$ . **b**, Triplon condensate in the superfluid phase. The hopping amplitude t of the quasiparticles leads to a uniform mixture of singlet and triplet states. **c**, Analogue of the Mott insulating phase for triplons with a magnetization plateau at  $m_z = 1/3$ : triplons 'crystallize' to form a superstructure. This phase is observed in  $SrCu_2(BO_3)_2$ , where quasiparticle hopping is strongly suppressed by geometrical frustration  $^{10.67,68,70}$ . **d**, Supersolid on a square lattice of dimers with exchange anisotropy as proposed theoretically  $^{89,91}$ , and for other lattice geometries  $^{86-88,90}$ . Both translation and U(1) symmetry are spontaneously broken, that is, the superfluid and 'crystal' of lattice bosons coexist.

# **BEYOND SIMPLE BEC**

The interplay of an atomic lattice and strong boson interactions leads to new phenomena beyond the simple BEC paradigm. When the boson density approaches a value commensurate with the lattice periodicity, we often find magnetization plateaux where the triplon density stays constant in a finite range of magnetic fields at T = 0. We have already discussed two simple examples of such plateaux for  $H < H_{c1}$  with zero magnetization and for  $H > H_{c2}$ where each site is occupied by one triplon (Fig. 1d). These simple  $m_z = 0$  or 1 plateaux correspond to states with an integer number of bosons per unit cell of the lattice. More generally, because in spin systems the  $S_{m,i}^z S_{n,j}^z$  terms in the hamiltonian (1) translate into triplon repulsion, magnetization plateaux at fractional fillings can also occur<sup>63,64</sup>. Such intermediate plateaux arise for strong enough repulsion between spin excitations on adjacent lattice sites and beyond, and correspond in boson language to Mott-insulator phases 10,65,66. In the simplest case (square lattice, repulsive nearestneighbour interactions) triplons may form an incompressible state by 'crystallizing' in a superstructure pattern (Fig. 3c). Unlike the ground state of an integer magnetization plateau (Fig. 3b), such a state violates the translational symmetry of the lattice, and the spontaneous symmetry breaking is characterized by a discrete order parameter. Fractional plateaux require strong magnon interactions (in comparison to the kinetic energy) and are therefore less robust then integer ones. Nonetheless, plateaux at magnon fillings 1/3, 1/4 and 1/8, and recently for other fractions have been observed in  $SrCu_2(BO_3)_2$  (refs 67–69). The kinetic energy in this material is suppressed by geometrical frustration and magnon repulsion is therefore more pronounced<sup>70</sup>.

Outside the plateaux, magnons can be easily added to the condensate, ensuring a continuously varying magnetization  $m_z(H)$ . This state has transverse magnetic order violating the continuous O(2) symmetry, is therefore gapless and corresponds to the superfluid phase of bosons. Its excitations are Goldstone modes, characteristics of this superfluid phase<sup>11,14</sup>. The incompressible plateau state and the gapless state with a continuously varying magnetization are again separated by a QCP.

Although the dimensionality of an antiferromagnet is determined mostly by quantum chemistry, it may also change on the fly as the magnet is cooled down. The 'Han purple' BaCuSi<sub>2</sub>O<sub>6</sub> contains layers of dimers with finite exchange couplings between the layers. Yet the critical behaviour shows two-dimensional exponents and an interesting crossover close to the QCP<sup>37,40</sup>. Attempts to interpret these surprising facts were made on the basis of ideas that the interplane triplon hopping is suppressed

by geometrical frustration and vanishes exactly at the wavevector of the condensate<sup>37,38</sup>. This would lead to a dimensional reduction at low temperature. An alternative explanation points to the existence of inequivalent layers with different values of the spin gap as the cause of the two-dimensional behaviour<sup>35,39,40</sup>.

Another spectacular example of reduced dimensionality is provided by (C<sub>5</sub>H<sub>12</sub>N)<sub>2</sub>CuBr<sub>4</sub> (refs 71,72) and related organometallic compounds. These materials can be thought of as a collection of chains of dimers nearly decoupled from one another. As a result, a large part of the H-T phase diagram can be understood by starting in the one-dimensional limit<sup>7,73–77</sup>. At low energies the triplons on a single chain form a Luttinger liquid<sup>78</sup>. Such a compound is thus particularly well suited for studying the dimensional crossover between one and three dimensions. In the former case, spin excitations behave as fermions, as described by the Jordan-Wigner transformation; in the latter, the excitations act like 'regular' bosons<sup>79</sup>. The excellent degree of control over the density by the magnetic field as well as the various probes providing access to the dynamical spin-spin correlations make quasi-one-dimensional magnets ideal for studying the dimensional crossover, a phenomenon of general importance for several major areas in physics<sup>78</sup>.

We would like to mention the tantalizing possibility of finding an exotic phase known as the supersolid80. A hybrid of a solid and a superfluid, this phase violates both the translational symmetry by forming a density wave and the U(1) phase symmetry by showing transverse magnetic order (Fig. 3d). It was conjectured long ago<sup>81–83</sup> and its existence is under intense debate at the moment following experiments in helium84,85. Such a phase could occur at the end of a fractional plateau because the transition must accomplish two tasks: create a superfluid and destroy a solid (that is, restore the broken translational symmetry). According to Landau's theory of phase transitions, such transitions are either discontinuous or happen in two stages with an intervening, possibly supersolid, phase. Several theoretical calculations show that supersolids exist in realistic magnetic models<sup>86–91</sup>. The observation of such a phase would thus be an interesting realization of the physics of interacting bosons on a lattice.

Last but not least, advances in growth techniques may enable us in the future to study in detail the effects of disorder on bosons<sup>66,92</sup>. Here Bose glasses, which can be made by adding bond disorder or site dilution, are of current interest<sup>66,93–96</sup>, as is the influence of defects on the quantum critical states<sup>97,98</sup>. Other directions include thermodynamics of strongly interacting bosons<sup>41,99</sup> and quantum coherence at the mesoscopic scale<sup>100</sup>.

# CONCLUSIONS AND OUTLOOK

Quantum spins and quantum dimer systems offer remarkable opportunities to study phenomena related to the Bose–Einstein condensation of interacting quantum particles. The high density and low mass of spin excitations, magnons or triplons, leads to robust condensates, which survive to temperatures as high as 10 K. The availability of numerous model magnets and physical probes has enabled a detailed study of the critical phenomena and magnetic properties in the vicinity of the BEC quantum phase transition. Spin systems have thus proven to be nicely complementary to other systems, such as cold atomic gases, for the investigation of the BEC. The success of this approach and present state of knowledge in the field is the result of a collective effort across several areas of expertise, including materials science and chemistry, where high-quality samples are prepared.

In addition to providing a direct connection with the remarkable BEC phenomenon, the boson picture offers an intuitive understanding for complex quantum properties of matter that are much less physically transparent in the original spin language and provides access to a cornucopia of exciting new problems and questions. For example, a classical description of the dimerized magnetic materials in the high-field ordered phase as canted antiferromagnets misses entirely the relevant spin physics close to the quantum phase transition. The transverse order vanishes at the OCP and all localized magnetic moments are in a highly fluctuating, perfectly entangled quantum-mechanical state, which finds a clear-cut and intuitive physical description in boson language. The possibility to fine-tune the density of bosons by using the magnetic field provides a tool to determine the role of the interactions in such systems. A wide range of different lattice geometries and dimensionalities is available for further study. In combination with the large number of experimental probes for spins, this enables us to enter the world of exotic phases of strongly interacting quantum particles such as the BEC in various dimensions, Luttinger-liquid physics, commensurate solids with a fractional number of bosons per unit cell and supersolids combining superfluidity with a broken translational symmetry.

#### doi:10.1038/nphys893

### References

- 1. Bose, S. N. Plancks Gesetz und Lichtquantenhypothese. Z. Phys. 26, 178–181 (1924).
- Einstein, A. Quantentheorie des einatomigen idealen Gases. Sitz. Ber. Kgl. Preuss. Akad. Wiss 261–267 (1924).
- Bloch, F. Zur Theorie des Ferromagnetismus, Z. Physik 61, 206–219 (1930).
- Matsubara, T. & Matsuda, H. A lattice model of liquid helium. Prog. Theor. Phys. 16, 569-582 (1956)
- Batyev, E. G. & Braginskii, L. S. Antiferromagnet in a strong magnetic field: Analogy with Bose gas Sov. Phys. JETP 60, 781–786 (1984).
- Affleck, I. Theory of Haldane-gap antiferromagnets in applied fields. Phys. Rev. B 41, 6697–6702 (1990).
- Giamarchi, T. & Tsvelik, A. M. Coupled ladders in a magnetic field. Phys. Rev. B 59, 11398–11407 (1999).
- Nikuni, T., Oshikawa, M., Oosawa, A. & Tanaka, H. Bose–Einstein condensation of diluted magnons in TlCuCl<sub>3</sub>. Phys. Rev. Lett. 84, 5868–5871 (2000).
- Wessel, S., Olshanii, M. & Haas, S. Field-induced magnetic order in quantum spin liquids. Phys. Rev. Lett. 87, 206407 (2001).
- 10. Rice, T. M. To condense or not to condense. Science 298, 760–761 (2002).
- Matsumoto, M., Normand, B., Rice, T. M. & Sigrist, M. Magnon dispersion in the field-induced magnetically ordered phase of TlCuCl<sub>3</sub>. Phys. Rev. Lett. 89, 077203 (2002).
- Matsumoto, M., Normand, B., Rice, T. M. & Sigrist, M. Field- and pressure-induced magnetic quantum phase transitions in TlCuCl<sub>3</sub>. Phys. Rev. B 69, 054423 (2004).
- Nohadani, O., Wessel, S., Normand, B. & Haas, S. Universal scaling at field-induced magnetic phase transitions. Phys. Rev. B 69, 220402 (2004).
- Rüegg, C. et al. Bose–Einstein condensation of the triplet states in the magnetic insulator TlCuCl<sub>3</sub> Nature 423, 62–65 (2003).
- Demokritov, S. O. et al. Bose–Einstein condensation of quasi-equilibrium magnons at room temperature under pumping. Nature 443, 430–433 (2006).
- 16. Kasprzak, J. et al. Bose–Einstein condensation of exciton polaritons. Nature 443, 409–414 (2006).
- Volovik, G. E. Twenty years of magnon Bose condensation and spin current superfluidity in <sup>3</sup>He-B. Preprint at <a href="http://arxiv.org/abs/cond-mat/0701180">http://arxiv.org/abs/cond-mat/0701180</a> (2007).
- Bunkov, Y. M. & Volovik, G. E. Magnon condensation into a Q ball in <sup>3</sup>He-B. Phys. Rev. Lett. 98, 265302 (2007).
- Nogueira, F. S. & Bennemann, K.-H. Spin Josephson effect in ferromagnet/ferromagnet tunnel junctions. Europhys. Lett. 67, 620–626 (2004).
- Syromyatnikov, A. V. Bose–Einstein condensation of magnons in magnets with predominant ferromagnetic interactions. *Phys. Rev. B* 75, 134421 (2007).
- 21. Pitaevskii, L. & Stringari, S. Bose-Einstein Condensation (Clarendon, Oxford, 2003).
- Greiner, M., Mandel, O., Esslinger, T., Hänsch, T. W. & Bloch, I. Quantum phase transition from a superfluid to a Mott insulator in a gas of ultracold atoms. *Nature* 415, 39–44 (2002).
- 23. Bloch, I. Ultracold quantum gases in optical lattices. *Nature Phys.* 1, 23–30 (2005).
- Bloch, I., Dalibard, J. & Zwerger, W. Many-body physics with ultracold gases. Rev. Mod. Phys. (in the press); preprint at <a href="http://arxiv.org/abs/cond-mat/0704.3011">http://arxiv.org/abs/cond-mat/0704.3011</a>> (2007).
- Cavadini, N. et al. Magnetic excitations in the quantum spin system TlCuCl<sub>3</sub>. Phys. Rev. B 63, 172414 (2001).
- Cavadini, N. et al. Triplet excitations in low-H<sub>ε</sub> spin gap systems KCuCl<sub>3</sub> and TlCuCl<sub>3</sub>: An inelastic neutron scattering study. Phys. Rev. B 65, 132415 (2002).
- Johannsen, N., Vasiliev, A., Oosawa, A., Tanaka, H. & Lorenz, T. Magnetoelastic coupling in the spin-dimer system TlCuCl<sub>3</sub>. Phys. Rev. Lett. 95, 017205 (2005).
- Vyaselev, O., Takigawa, M., Vasiliev, A., Oosawa, A. & Tanaka, H. Field-induced magnetic order and simultaneous lattice deformation in TlCuCl., Phys. Rev. Lett. 92, 207202 (2004).
   Sherman, E. Y., Lemmens, P., Buses, B., Oosawa, A. & Tanaka, H. Sound attenuation study on the
- Sherman, E. Y., Lemmens, P., Busse, B., Oosawa, A. & Tanaka, H. Sound attenuation study on the Bose–Einstein condensation of magnons in TlCuCl<sub>3</sub>. Phys. Rev. Lett. 91, 057201 (2003).
- Misguich, G. & Oshikawa, M. Bose–Einstein condensation of magnons in TlCuCl<sub>3</sub>: Phase diagram
  and specific heat from a self-consistent Hartree–Fock calculation with a realistic dispersion relation.
   J. Phys. Soc. Japan 73, 3429–3434 (2004).
- Glazkov, V. N., Smirnov, A. I., Tanaka, H. & Oosawa, A. Spin-resonance modes of the spin-gap magnet TlCuCl<sub>3</sub>. Phys. Rev. B 69, 184410 (2004).
- Kolezhuk, A. K., Glazkov, V. N., Tanaka, H. & Oosawa, A. Dynamics of an anisotropic spin dimer system in a strong magnetic field. *Phys. Rev. B* 70, 020403 (2004).
- Sirker, J., Weisse, A. & Sushkov, O. P. Consequences of spin-orbit coupling for the Bose-Einstein condensation of magnons. Europhys. Lett. 68, 275–281 (2004).

# REVIEW ARTICLE | FOCUS

- Sasago, Y., Uchinokura, K., Zheludev, A. & Shirane, G. Temperature-dependent spin gap and singlet ground state in BaCuSi<sub>2</sub>O<sub>6</sub>. Phys. Rev. B 55, 8357–8360 (1997).
- Rüegg, C. et al. Multiple magnon modes and consequences for the Bose–Einstein condensed phase in BaCuSi<sub>2</sub>O<sub>6</sub>. Phys. Rev. Lett. 98, 017202 (2007).
- Jaime, M. et al. Magnetic-field-induced condensation of triplons in Han Purple pigment BaCuSi<sub>2</sub>O<sub>6</sub>. Phys. Rev. Lett. 93, 087203 (2004).
- Sebastian, S. E. et al. Dimensional reduction at a quantum critical point. Nature 441, 617–620 (2006).
- Batista, C. D. et al. Geometric frustration and dimensional reduction at a quantum critical point. Phys. Rev. Lett. 98, 257201 (2007).
- Rösch, O. & Vojta, M. Reduced dimensionality in layered quantum dimer magnets: Frustration vs. inhomogeneous condensates. Phys. Rev. B 76, 224408 (2007).
- Krämer, S. et al. Nuclear magnetic resonance evidence for a strong modulation of the Bose–Einstein condensate in BaCuSi-Oc. Phys. Rev. B 76, 100406 (2007).
- condensate in BaCuSi<sub>2</sub>O<sub>6</sub>. *Phys. Rev. B* 76, 100406 (2007).
   Xu, G. Y., Broholm, C., Reich, D. H. & Adams, M. A. Triplet waves in a quantum spin liquid. *Phys. Rev. Lett.* 84, 4465–4468 (2000).
- 42. Shiramura, W. *et al.* Magnetisation plateaus in NH<sub>4</sub>CuCl<sub>3</sub>. *J. Phys. Soc. Japan* **67**, 1548–1551 (1998).
- Rüegg, C. et al. Neutron scattering study of the field-dependent ground state and the spin dynamics in spin-one-half NH<sub>4</sub>CuCl<sub>3</sub>. Phys. Rev. Lett. 93, 037207 (2004).
- Matsumoto, M. Microscopic model for the magnetisation plateaus in NH<sub>4</sub>CuCl<sub>3</sub>. Phys. Rev. B 68, 180403 (2003).
- Grenier, B. et al. Spin excitations throughout the field-induced magnetic phase of the spin 1/2 alternating chain compound Cu(NO<sub>3</sub>)<sub>2</sub> · 2.5D<sub>2</sub>O, J. Magn. Magn. Matter 310, 1269–1271 (2007).
   Grenier, B. et al. Ordering and excitations in the field-induced magnetic phase of Cs<sub>C</sub>Cr<sub>3</sub>B<sub>1</sub>, Phys.
- Grenier, B. et al. Ordering and excitations in the field-induced magnetic phase of Cs<sub>3</sub>Cr<sub>2</sub>Br<sub>9</sub>. Phys Rev. Lett. 92, 177202 (2004).
- Garlea, V. O. et al. Excitations from a Bose–Einstein condensate of magnons in coupled spin ladders. Phys. Rev. Lett. 98, 167202 (2007)
- Masuda, T. et al. Dynamics of composite Haldane spin chains in IPA-CuCl<sub>3</sub>. Phys. Rev. Lett. 96, 047210 (2006).
- 49. Stone, M. B. et al. Field-driven phase transitions in a quasi-two-dimensional quantum antiferromagnet. New I. Phys. 9, 31 (2007)
- antiferromagnet. New J. Phys. 9, 31 (2007).
  50. Zheludev, A. et al. Quasielastic neutron scattering in the high-field phase of a Haldane antiferromagnet. Phys. Rev. Lett. 88, 077206 (2002).
- Zapf, V. S. et al. Bose–Einstein condensation of S = 1 nickel spin degrees of freedom in NiCl<sub>2</sub>-4SC (NH<sub>2</sub>)<sub>2</sub>. Phys. Rev. Lett. 96, 077204 (2006).
- Zvyagin, S. A. et al. Magnetic excitations in the spin-1 anisotropic Heisenberg antiferromagnetic chain system NiCl<sub>2</sub>-4SC (NH<sub>2</sub>)<sub>2</sub>. Phys. Rev. Lett. 98, 047205 (2007).
- Coldea, R. et al. Direct measurement of the spin hamiltonian and observation of condensation of magnons in the 2D frustrated quantum magnet Cs<sub>2</sub>CuCl<sub>4</sub>. Phys. Rev. Lett. 88, 137203 (2002).
- Radu, T. et al. Bose–Einstein condensation of magnons in Cs<sub>2</sub>CuCl<sub>4</sub>. Phys. Rev. Lett. 95, 127202 (2005).
- Tachiki, M. & Yamada, T. Spin ordering in a spin-pair system. J. Phys. Soc. Japan 28, 1413–1425 (1970).
- Maeda, Y., Hotta, C. & Oshikawa, M. Universal temperature dependence of the magnetisation of gapped spin chains. *Phys. Rev. Lett.* 99, 057205 (2007).
- Sebastian, S. E. et al. Role of anisotropy in the spin-dimer compound BaCuSi<sub>2</sub>O<sub>6</sub>. Phys. Rev. B 74, 180401 (2006).
- Clémancey, M. et al. Field-induced staggered magnetization and magnetic ordering in Cu<sub>2</sub> (C<sub>5</sub>H<sub>12</sub>N<sub>2</sub>)<sub>2</sub>Cl<sub>4</sub>. Phys. Rev. Lett. 97, 167204 (2006).
- Oshikawa, M. & Affleck, I. Field-induced gap in S = 1/2 antiferromagnetic chains. Phys. Rev. Lett. 79, 2883–2886 (1997).
- Miyahara, S. et al. Uniform and staggered magnetizations induced by Dzyaloshinskii–Moriya interactions in isolated and coupled spin-1/2 dimers in a magnetic field. Phys. Rev. B 75, 184402 (2007).
- Matsumoto, M. & Sigrist, M. Ehrenfest relations and magnetoelastic effects in field-induced ordered phases. J. Phys. Soc. Japan 74, 2310–2316 (2005).
- 62. Orignac, E. & Citro, R. Magnetostriction in an array of spin chains under a magnetic field. *Phys. Rev. B* 71, 214419 (2005).
- Oshikawa, M., Yamanaka, M. & Affleck, I. Magnetisation plateaus in spin chains: 'Haldane gap' for half-integer spins. Phys. Rev. Lett. 78, 1984–1987 (1997).
- Cabra, D. C., Honecker, A. & Pujol, P. Magnetisation plateaux in N-leg spin ladders. Phys. Rev. B 58, 6241–6257 (1998).
- Haldane, F. D. M. Effective harmonic-fluid approach to low-energy properties of one-dimensional quantum fluids. *Phys. Rev. Lett.* 47, 1840–1843 (1981).
   Fisher, M. P. A., Weichman, P. B., Grinstein, G. & Fisher, D. S. Boson localization and the
- Fisher, M. P. A., Weichman, P. B., Grinstein, G. & Fisher, D. S. Boson localization and the superfluid–insulator transition. *Phys. Rev. B* 40, 546–570 (1989).
- Kageyama, H. et al. Exact dimer ground state and quantized magnetization plateaus in the two-dimensional spin system SrCu<sub>2</sub>(BO<sub>3</sub>)<sub>2</sub>. Phys. Rev. Lett. 82, 3168–3171 (1999).

- Kodama, K. et al. Magnetic superstructure in the two-dimensional quantum antiferromagnet SrCu<sub>2</sub>(BO<sub>3</sub>). Science 298, 395–399 (2002).
- Sebastian, S. E. et al. Fractalization drives crystalline states in a frustrated spin system. Preprint at <a href="http://arxiv.org/abs/cond-mat/0707.2075">http://arxiv.org/abs/cond-mat/0707.2075</a>> (2007).
- Miyahara, S. & Ueda, K. Theory of the orthogonal dimer Heisenberg spin model for SrCu<sub>2</sub>(BO<sub>3</sub>)<sub>2</sub>.
   J. Phys. Condens. Matter 15, R327–R366 (2003).
- Watson, B. C. et al. Magnetic spin ladder (C<sub>5</sub>H<sub>12</sub>N)<sub>2</sub>CuBr<sub>4</sub>: High-field magnetisation and scaling near quantum criticality. Phys. Rev. Lett. 86, 5168–5171 (2001).
- Lorenz, T. et al. Diverging thermal expansion of the spin-ladder system (C<sub>3</sub>H<sub>12</sub>N)<sub>2</sub>CuBr<sub>4</sub>. Phys. Rev. Lett. (in the press); preprint at <a href="http://arxiv.org/abs/cond-mat/0711.1510">http://arxiv.org/abs/cond-mat/0711.1510</a> (2007).
- Chitra, R. & Giamarchi, T. Critical properties of gapped spin-1/2 chains and ladders in a magnetic field. Phys. Rev. B 55, 5816–5826 (1997).
- Mila, F. Ladders in a magnetic field: A strong coupling approach. Eur. Phys. J. B 6, 201–205 (1998).
- Furusaki, A. & Zhang, S.-C. Dynamical spin correlations in the Heisenberg ladder under a magnetic field and correlation functions in the SO(5) ladder. *Phys. Rev. B* 60, 1175–1187 (1999).
- Usami, M. & Suga, S. Critical properties of S = 1/2 Heisenberg ladders in magnetic fields. *Phys. Rev. B* 58, 14401–14406 (1998).
- Hikihara, T. & Furusaki, A. Spin correlations in the two-leg antiferromagnetic ladder in a magnetic field. Phys. Rev. B 63, 134438 (2001).
- 78. Giamarchi, T. Quantum Physics in One Dimension (Oxford Univ. Press, Oxford, 2004).
- Orignac, E., Citro, R. & Giamarchi, T. Critical properties and Bose–Einstein condensation in dimer spin systems. Phys. Rev. B 75, 140403 (2007).
- 30. Prokof'ev, N. & Svistunov, B. Supersolid state of matter. Phys. Rev. Lett. 94, 155302 (2005).
- Andreev, A. F. & Lifshitz, I. M. Quantum theory of defects in crystals. Sov. Phys. JETP 29, 1107–1113 (1969).
- Chester, G. V. Speculations on Bose–Einstein condensation and quantum crystals. Phys. Rev. A 2, 256–258 (1970).
- 83. Leggett, A. J. Can a solid be superfluid?. Phys. Rev. Lett. 25, 1543-1546 (1970).
- Kim, E. & Chan, M. H. W. Probable observation of a supersolid helium phase. Nature 427, 225–227 (2004).
- Sasaki, S., Ishiguro, R., Caupin, F., Maris, H. J. & Balibar, S. Superfluidity of grain boundaries and supersolid behaviour. Science 313, 1098–1100 (2006).
- Wessel, S. & Troyer, M. Supersolid hard-core bosons on the triangular lattice. *Phys. Rev. Lett.* 95, 127205 (2005).
- Heidarian, D. & Damle, K. Persistent supersolid phase of hard-core bosons on the triangular lattice. Phys. Rev. Lett. 95, 127206 (2005).
- Melko, R. G. et al. Supersolid order from disorder: Hard-core bosons on the triangular lattice. Phys. Rev. Lett. 95, 127207 (2005).
- Ng, K. K. & Lee, T. K. Supersolid phase in spin dimer XXZ systems under a magnetic field. Phys. Rev. Lett. 97, 127204 (2006).
- Sengupta, P. & Batista, C. D. Field-induced supersolid phase in spin-one Heisenberg models. *Phys. Rev. Lett.* 98, 227201 (2007).
- Laflorencie, N. & Mila, F. Quantum and thermal transitions out of the supersolid phase of a 2D quantum antiferromagnet. Phys. Rev. Lett. 99, 027202 (2007).
- Giamarchi, T. & Schulz, H. J. Anderson localization and interactions in one-dimensional metals. Phys. Rev. B 37, 325–340 (1988).
- Nohadani, O., Wessel, S. & Haas, S. Bose-glass phases in disordered quantum magnets. Phys. Rev. Lett. 95, 227201 (2005).
- 94. Roscilde, T. & Haas, S. Quantum localization in bilayer Heisenberg antiferromagnets with site dilution. *Phys. Rev. Lett.* **95**, 207206 (2005).
- Yu, R., Roscilde, T. & Haas, S. Quantum disorder and Griffiths singularities in bond-diluted two-dimensional Heisenberg antiferromagnets. *Phys. Rev. B* 73, 064406 (2006).
- two-dimensional Heisenberg antiferromagnets. *Phys. Rev. B* **73**, 064406 (2006).

  96. Roscilde, T. Field-induced quantum-disordered phases in S = 1/2 weakly coupled dimer systems with site dilution. *Phys. Rev. B* **74**, 144418 (2006).
- 97. Xu, G. *et al.* Holes in a quantum spin liquid. *Science* **289**, 419–422 (2000).
- Roscilde, T. & Haas, S. Mott glass in site-diluted S = 1 antiferromagnets with single-ion anisotropy. Phys. Rev. Lett. 99, 047205 (2007).
- Rüegg, C. et al. Quantum statistics of interacting dimer spin systems. Phys. Rev. Lett. 95, 267201 (2005).
- 100. Xu, G. et al. Mesoscopic phase coherence in a quantum spin fluid. Science 317, 1049–1052 (2007).

## Acknowledgements

This work was supported in part by the Swiss NSF under NCCR MaNEP, a Wolfson Royal Society Research Merit Award and the US National Science Foundation.

Correspondence and requests for materials should be addressed to T.G., C.R. or O.T.

Reprints and permission information is available online at http://npg.nature.com/reprintsandpermissions/