## Chapter 10

# Magnetization plateaus

A standard way to probe magnetic systems is to measure their response to a magnetic field. In gapped magnets, and more generally in frustrated quantum magnets, it is very instructive to keep track of the full magnetization curve up to the saturation field (if at all possible!), and not just of the slope at zero field, the magnetic susceptibility. At zero temperature, all gapped antiferromagnets have a plateau at zero magnetization up to the field that brings one of the triplets below the singlet, the prominent example being the spin-1 chain. When frustration is present, other magnetization plateaus can appear at fractional values of the magnetization, even if the system is gapless at zero field, as e.g. the triangular lattice Heisenberg model. In this chapter, I review the two main mechanisms that have been identified, the stabilization of a collinear classical ground state in a field range by an order by disorder mechanism, and the stabilization of Wigner crystals of magnetic excitations in dimer based frustrated magnets.

## 10.1 Introduction

The Hamiltonian of a Heisenberg magnet in a uniform field  $\vec{H}$  is given by

$$\mathcal{H} = \sum_{(i,j)} J_{ij} \vec{S}_i \cdot \vec{S}_j - \vec{H} \cdot \sum_i \vec{S}_i$$
 (10.1)

where the g-factor and the Bohr magneton  $\mu_B$  have been incorporated in the field  $\vec{H}$ . Very often it will be useful to choose a quantization axis. The natural choice is to choose that of the field. Without loss of generality, the field can be chosen along z,  $\vec{H} = H\hat{z}$ , and the Hamiltonian reads

$$\mathcal{H} = \sum_{(i,j)} J_{ij} \vec{S}_i \cdot \vec{S}_j - H \sum_i S_i^z. \tag{10.2}$$

When the intensity of the field is large enough, larger than a critical field known as the saturation field  $H_{\rm sat}$ , all spins point along  $\vec{H}$ , and the ground state is a simple product state. Below that field, the configuration is the result of a compromise between exchange interactions, which tend to align spins anti-parallel to each other if they are antiferromagnetic, and the magnetic field, which tends to align spins parallel to itself. The outcome of this competition depends dramatically on many parameters (nature of the spins, classical or quantum, value of S for quantum spins, topology of the lattice, temperature), with a magnetization curve that is simply linear for many classical systems at zero temperature but can develop several anomalies for highly frustrated quantum systems.

## 10.2 Semi-classical plateaus: Triangular lattice

Certain plateaus correspond to classical configurations of spin. The prominent example is the 1/3 plateau of the triangular lattice. In that case, thermal fluctuations or quantum fluctuations have been shown to stabilize a plateau at 1/3 of the magnetization. Let us see the underlying mechanism.

To start, let us discuss the classical ground state. For that purpose, it is useful to rewrite the energy as a sum over triangles:

$$E = \sum_{i} E_{i}$$

with

$$E_i = \frac{J}{2} (\vec{S}_{i_1}.\vec{S}_{i_2} + \vec{S}_{i_2}.\vec{S}_{i_3} + \vec{S}_{i_3}.\vec{S}_{i_1}) - \frac{1}{6} \vec{H}.(\vec{S}_{i_1} + \vec{S}_{i_2} + \vec{S}_{i_3})$$

where the sum over i runs over all triangles, and where  $i_1$ ,  $i_2$  and  $i_3$  are the corners of triangle i. The coupling constant J appears with a factor 1/2 because each bond belongs to 2 triangles, and the field with a factor 1/6 because each spin belongs to 6 triangles.

 $E_i$  can be rewritten in terms of the total spin as

$$E_{i} = \frac{J}{4} \left[ (\vec{S}_{i_{1}} + \vec{S}_{i_{2}} + \vec{S}_{i_{3}})^{2} - 3 \right] - \frac{1}{6} \vec{H} \cdot (\vec{S}_{i_{1}} + \vec{S}_{i_{2}} + \vec{S}_{i_{3}})$$

$$= \frac{J}{4} \left( \vec{S}_{\text{tot}}(i)^{2} - \frac{2\vec{H}}{3J} \cdot \vec{S}_{\text{tot}}(i) \right) - \frac{3J}{4}$$

$$= \frac{J}{4} \left( \vec{S}_{\text{tot}}(i) - \frac{\vec{H}}{3J} \right)^{2} - \frac{J}{4} \frac{H^{2}}{9J^{2}} - \frac{3J}{4}$$

The energy of a triangle will thus be minimal as soon as  $\vec{S}_{\text{tot}}(i) - \frac{\vec{H}}{3J}$ , a condition that can be fulfilled up to H = 9J since  $\vec{S}_{\text{tot}}(i)$  is the sum of three spins of length 1. This condition can then be satisfied in many ways, for instance in an umbrella state or a coplanar state (see Fig. 10.1), and this condition can be simultaneously satisfied in all triangles by adopting a three-sublattice structure.

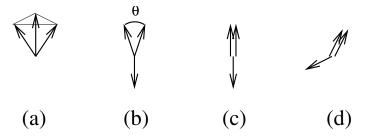


Figure 10.1: Some classical ground states of the triangular lattice Heisenberg antiferromagnet in a field. (a) An umbrella configuration; (b) Coplanar ground state for  $H < H_{\text{sat}}/3$ ; (c) Collinear ground state for  $H = H_{\text{sat}}/3$ ; (d) Coplanar ground state for  $H > H_{\text{sat}}/3$ .

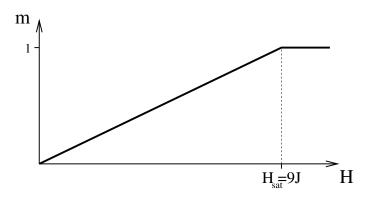


Figure 10.2: Zero-temperature magnetization of the classical triangular lattice antiferromagnet.

So, at zero temperature, the magnetization per spin is given by  $\vec{m} = \frac{\vec{H}}{9J}$ . This is valid up to the saturation field  $H_{\rm sat} = 9J$  beyond which  $\vec{m} = \vec{H}/H$ . The classical magnetization thus grows linearly with the field at zero temperature (see Fig. 10.2).

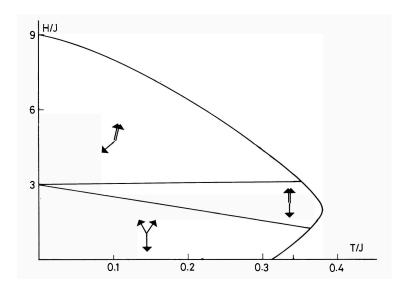


Figure 10.3: Temperature-field phase diagram of the classical triangular lattice antiferromagnet (after Kawamura and Miyashita, 1985).

However, the fact that the condition  $\vec{S}_{\rm tot}(i) = \frac{\vec{H}}{3J}$  leads to a huge degeneracy suggests that we are in a situation where thermal or quantum fluctuations might pick one type of configuration and change the physics. And indeed, the coplanar configuration is stabilized over the non-coplanar configurations. However, this is not the whole story. At  $\vec{H} = \vec{H}_{\rm sat}/3$ , the configuration is collinear, with two spins along the field and one spin opposite to the field. One can expect this configuration to be stabilized over a certain field range.

## 10.2.1 Thermal fluctuations

This has been verified with Monte Carlo simulations by Kawamura and Miyashita, who found numerically that, at finite temperature, the collinear structure is indeed stabilized in a finite field range (see Fig. 10.3).

### 10.2.2 Quantum fluctuations

As usual, one expects the same to be true with quantum fluctuations. The argument is more subtle however than with degenerate ground states because, if a plateau is stabilized, it means that a type of order is stabilized in

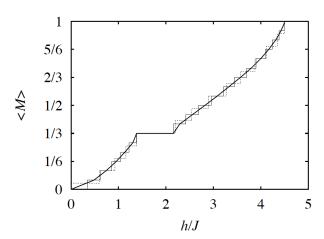


Figure 10.4: Magnetization curve of the S=1/2 Heisenberg antiferromagnet on the triangular lattice. The thin dashed and solid line are for N=27 and N=36 sites, respectively. The bold line is an extrapolation to the thermodynamic limit (after Honecker et al, 2004).

a region where the classical order is unstable.

How is this possible? Obviously, this requires to push the spin-wave expansion to higher-order, a rather cumbersome task. Fortunately, the stabilization of the 1/3 plateau can be understood qualitatively in a simple way. The starting point is that, in helical systems, where the structure depends on a parameter (here the magnetic field) the structure is influenced by quantum fluctuations. In the present case, the angle  $\theta$  of the structure below  $H_{\text{sat}}/3$  (see Fig.10.1) is not the same for classical spins and for finite S. So the critical field  $H_{c_1}$  where it becomes collinear will have 1/S corrections. The same is true coming from above, and the critical field  $H_{c_2}$  where it becomes collinear does not need to be equal to  $H_{c_1}$ .

Now, a simple argument suggests that  $H_{c_1} < H_c < H_{c_2}$ . Indeed, for a given field  $H < H_c$ , the coplanar classical ground state is defined by  $\theta_{\rm cl}$ , which satisfies the minimality condition:

$$\left. \frac{\partial E_{\rm cl}}{\partial \theta} \right|_{\theta = \theta_{\rm cl}} = 0 \tag{10.3}$$

As  $H \to H_c = \frac{H_{\text{sat}}}{3}$ ,  $\theta_{\text{cl}} \to 0$ . But quantum corrections favour small angles. So,  $\theta_{\text{quan}} < \theta_{\text{cl}}$ , and one can expect  $H_{c_1} < H_c$ . The same reasoning leads

to  $H_{c_2} > H_c$ . Pushing the 1/S expansion to higher order, Chubukov has actually calculated these critical fields, and he found:

$$\begin{cases}
H_{c_1} = H_c \left(1 - \frac{0.084}{S}\right) \\
H_{c_2} = H_c \left(1 + \frac{0.215}{S}\right)
\end{cases}$$
(10.4)

This prediction of a 1/3-plateau for the triangular lattice has been confirmed for spins-1/2 by exact diagonalizations of finite clusters (see Fig. 10.4).

## 10.3 Quantum plateaux

The simplest example of a quantum plateau is the plateau that occurs in all gapped antiferromagnets. If we denote by  $\Delta$  the gap to the first triplet excitation, the relative energy of the triplet with  $S_{\rm tot}^z=1$  with respect to the singlet ground state is given by  $E(S_{\rm tot}^z=1,H)-E_{\rm GS}=\Delta-H$ . This energy vanishes at  $H_c=\Delta$ , signalling a phase transition into a phase with a finite magnetization. Up to that field, the ground state is unaffected by the field, and the magnetization is strictly equal to 0. This behaviour is not specific to frustrated magnets. It occurs for instance in the spin-1 chain, with its famous Haldane gap, in spin-1/2 ladders, or more generally in dimerized Heisenberg models.

However, when frustration is present in a dimer based model, the motion of the triplet can be severely affected, and the reduction of the kinetic energy combined with the repulsion between triplets due to the antiferromagnetic interactions can lead to incompressible phases at intermediate magnetization analogous to the Mott insulating phases of strongly correlated electrons. In the rest of this chapter, we will treat in some detail the case of the frustrated ladder, and we will discuss briefly the case of the Shastry-Sutherland model, where this simple mechanism explains some of the plateaus.

## 10.3.1 Isolated dimer

Let us start by the trivial but instructive case of an isolated spin-1/2 dimer in a field described by the Hamiltonian

$$\mathcal{H} = J\vec{S}_1 \cdot \vec{S}_2 - H(S_1^z + S_2^z)$$

The eigenstates of that problem are the singlet  $|S\rangle$ , of energy  $E_{S=0} = -3J/4$ , and the triplets:  $|T_1\rangle$ , of energy  $E_1 = J/4 - H$ ,  $|T_0\rangle$ , of energy  $E_0 = J/4$ , and

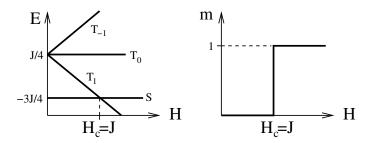


Figure 10.5: Left: Energy levels of a spin-1/2 dimer in a field. Right: Magnetization curve of a spin-1/2 dimer in a field.

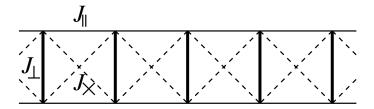


Figure 10.6: Sketch of the frustrated ladder.

 $|T_{-1}\rangle$ , of energy  $E_{-1} = J/4 + H$  (see Fig. 10.5, left panel). There is a phase transition at H = J, and the magnetization per site jumps from 0 to 1 in units of the magnetization per site at saturation (see Fig. 10.5, right panel).

## 10.3.2 Frustrated ladder

The spin-1/2 ladder is arguably the simplest example of a gapped quantum magnet. In the strong-rung limit, the calculation performed for the dimerized square lattice can be extended to that case and shows that the gap is of the order of the rung coupling up to a small correction due to the leg coupling. By contrast to the case of the dimerized square lattice however, field theory arguments show that the gap persists down to infinitesimal rung coupling.

In this section, we look at the effect of frustration by considering the frustrated ladder with rung coupling  $J_{\perp} > 0$ , and two types of inter-rung couplings between nearest-neighboring rungs,  $J_{\parallel} > 0$  between sites belonging to the same leg, and  $J_{\times} > 0$  between sites belonging to different legs, as shown

in Fig. 10.6. This model is defined by the Hamiltonian:

$$\mathcal{H} = J_{\perp} \sum_{i} \vec{S}_{i,1} \cdot \vec{S}_{i,2} + J_{\parallel} \sum_{i,l} \vec{S}_{i,l} \cdot \vec{S}_{i+1,l} + J_{\times} \sum_{i} (\vec{S}_{i,1} \cdot \vec{S}_{i+1,2} + \vec{S}_{i+1,1} \cdot \vec{S}_{i,2}) - H \sum_{i,l} S_{i,l}^{z},$$

where i keeps track of the position of the rung, l=1 or 2 denotes the leg, and  $\vec{S}_{i,l}$  stands for the spin that belongs to rung i and leg l. The presence of  $J_{\times}$  introduces odd loops, hence frustration.

#### Fully frustrated ladder

Let's start with the case of the fully frustrated ladder defined by  $J_{\parallel} = J_{\times} \equiv J$ . In that case, the model can be rewritten in terms of the total spins of the rungs  $\vec{T}_i = \vec{S}_{i,1} + \vec{S}_{i,2}$  as

$$\mathcal{H} = J \sum_{i} \vec{T}_{i} \cdot \vec{T}_{i+1} - H \sum_{i} T_{i}^{z} + \frac{J_{\perp}}{2} \sum_{i} (\vec{T}_{i}^{2} - \frac{3}{2}).$$

The Hilbert space can be decomposed into  $2^L$  sectors, where L is the number of rungs, according to the value of the total spin of each rung, which can be in a singlet or in a triplet state.

By arguments similar to those used in chapter 7 on gapped magnets, it is clear that the product of singlets on the rungs is an eigenstate, and that for large enough  $J_{\perp}/J$ , it will be the ground state in zero field. Upon reducing this ratio, there is a strong first order transition in zero field where all rungs become triplets. The contribution of the J term to the ground state energy is the ground state energy of the spin 1 chain, -1.401484...J per spin, and the transition occurs at  $J_{\perp}/J = 1.401484...$ 

We assume from now on that  $J_{\perp}/J$  is large enough for the product of singlets to be the ground state. The effect of the magnetic field can be simply understood in terms of the isolated dimer case. Indeed, the state where one rung is in a triplet state is still an eigenstate because it is coupled to singlets on both sides, and its energy will become lower than that of the singlet when  $H = J_{\perp}$ . However the state with only one triplet is not the ground state for  $H > J_{\perp}$ . Indeed, energy can be gained by turning other singlets into triplets as long as triplets are surrounded by singlets. The maximal energy will be gained when the maximal number of triplets surrounded by singlets is created, i.e. when every other rung is in a triplet state. There are two such states, with triplets occupying even or odd rungs, and their magnetization per site is equal to m = 1/2. The energy of that state as compared to the singlet state is given by

$$E_{m=1/2} - E_{S_{\text{tot}}=0} = (J_{\perp} - H) \frac{L}{2}.$$

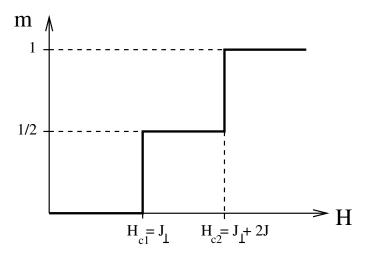


Figure 10.7: Magnetization curve of the fully frustrated ladder.

So there is a critical field  $H_{c1} = J_{\perp}$  where the magnetization jumps from 0 to half the saturation value.

Now, for large enough field, the ground state will be the fully polarized state with magnetization per site m = 1, and with energy

$$E_{m=1} - E_{S_{\text{tot}}=0} = (J_{\perp} - H)L + JL$$

where the second term comes from the exchange term J. This energy will be the ground state down to the field where it will be more advantageous to convert one triplet into a singlet surrounded by two triplets. But then, one will gain more energy by converting as many triplets as possible as long as the singlets are surrounded by two triplets, leading again to one of the two states with alternating singlets and triplets. The transition thus occurs when  $E_{m=1/2} = E_{m=1}$ , leading to a critical field  $H_{c2} = J_{\perp} + 2J$ . At that field, the magnetization jumps from half the saturation value to full saturation (see Fig. 10.7).

To summarize, the magnetization consists of a plateau at m = 0 up to  $H_{c1} = J_{\perp}$ , a plateau at m = 1/2 between  $H_{c1}$  and  $H_{c2} = J_{\perp} + 2J$ , and a plateau at saturation m = 1 above  $H_{c2}$ . The width of the plateau is equal to twice the inter-rung coupling and vanishes in the limit of isolated dimers, as it should.

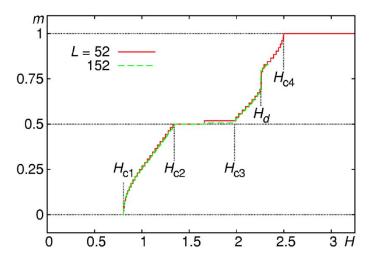


Figure 10.8: Magnetization curve of the frustrated ladder with  $J_{\parallel}/J_{\perp} = 0.55$  and  $J_{\times}/J_{\perp} = 0.7$  obtained with density matrix renormalization group (after Fouet et al, 2006).

## Partially frustrated ladder

How frustrated should the ladder be to support a 1/2 plateau? A simple answer can be given in the limit  $J_{\parallel}, J_{\times} \ll J_{\perp}$ . In that limit, the hopping of a triplet can be calculated using first-order perturbation theory, as we did in chapter 7 for the dimerized Heisenberg model on the square lattice. If we choose the same orientation for all rung singlets, say from leg l=1 to leg l=2, each cross couplings between two rungs leads to a hopping amplitudes  $-J_{\times}/4$ , adding to  $-J_{\times}/2$ , while each leg couplings leads to a hopping amplitude  $J_{\parallel}/4$ , adding to  $J_{\parallel}/2$ . So, in tight-binding language with hopping amplitude t, the triplet hopping is given by  $t=(J_{\parallel}-J_{\times})/2$ .

Now, triplets experience repulsion when they sit on neighboring sites because they have to pay the exchange energy of the antiferromagnetic couplings. Each exchange contributes with a factor 1/4, leading to an effective repulsion  $v = (J_X + J_{\parallel})/2$  between nearest neighbors.

Finally, the chemical potential is given by  $\mu = H - J_{\perp}$  since the energy of a triplet with respect to a singlet is equal to  $J_{\perp} - H$ , and by definition of the chemical potential this energy is equal to  $-\mu$ .

So the Hamiltonian describing the triplets can be reformulated as a spin-

less fermion Hamiltonian given by

$$\mathcal{H} = t \sum_{i} (c_i^{\dagger} c_{i+1}^{\phantom{\dagger}} + c_{i+1}^{\dagger} c_i^{\phantom{\dagger}}) + v \sum_{i} n_i n_{i+1} - \mu \sum_{i} n_i$$

where  $n_i = c_i^{\dagger} c_i$ , with  $t = (J_{\parallel} - J_{\times})/2$ ,  $v = (J_{\times} + J_{\parallel})/2$  and  $\mu = H - J_{\perp}$ . Now, this model has a Bethe ansatz solution, and there is a phase transition at v = 2t. Beyond that value, a gap opens at half-filling, and the ground state is a charge density wave of period 2. The condition v = 2t translates into  $J_{\times} = J_{\parallel}/3$ . So, in the limit of strong rungs, there will be a plateau at m = 1/2 if  $J_{\times}/J_{\parallel} > 1/3$ . In other words, it takes a finite level of frustration to develop a 1/2-plateau.

On the basis of these arguments, one could expect the generic curve of the frustrated ladder for  $1/3 < J_{\times}/J_{\parallel} < 1$  to have 4 critical fields: the field where the plateau at m = 0 ends, the beginning of the 1/2-plateau, the end of the 1/2-plateau, and the saturation field. In the limit  $J_{\times}/J_{\parallel} \rightarrow$ 1/3, the 1/2-plateau disappears, and one is left with two critical fields, the end of the plateau at m = 0, and the saturation field. In the opposite limit  $J_{\times}/J_{\parallel} \to 1$ , the intermediate regions between m=0 and m=1/2 and between m = 1/2 and m = 1 shrink to 0, and we are again left with only two critical fields at which the magnetization jumps. The actual situation is slightly more complicated (see Fig. 10.8). The fermionic model only describes singlets and triplets  $T_1$ . But in a ladder, there are also  $T_0$  and  $T_{-1}$ . It turns out that, when coming from saturation, and for some sets of parameters, it is more favourable to first create  $T_0$  triplets, resulting in another critical field between the end of the 1/2-plateau and saturation at which the magnetization jumps. Below this critical field, the ground state essentially consists of singlets and triplets  $T_1$  while above it, it is built out of  $T_0$  and  $T_1$  triplets.

## 10.3.3 Shastry-Sutherland model

The magnetization curve of the Shastry-Sutherland model is the richest observed so far in a frustrated quantum magnet, with plateaus at 1/8, 2/15, 1/6, 1/4, 1/3, 2/5 and 1/2 for a ratio of inter- to intra-dimer coupling of 0.63 according to steady and pulsed field experiments performed on a very accurate realization of that model in a spin-1/2 cuprate, SrCu<sub>2</sub>(BO<sub>3</sub>)<sub>2</sub>. The plateaus at 1/4, 1/3, 2/5 and 1/2 are crystals of triplets, and they can be understood as the result of a very small kinetic energy and long range repulsion between triplets. The lower magnetization plateaus however have another structure. Because of correlated hopping, a spin-2 bound state is

stabilized in zero field, and it has been predicted that it is the crystallization of such bound states that stabilizes the plateaus at 1/8, 2/15 and 1/6.