13 Quantum Magnetism In and Out of Equilibrium

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1 Quantum magnetism and strongly correlated systems

An exceptional and widely known quantum state of matter is superconductivity. It is realized by the spontaneous breaking of a (gauge) symmetry, in which (in the simplest theory) the interaction between the electrons and lattice degrees of freedom leads to this peculiar 'macroscopic' behavior of certain materials. Other systems, in which the interaction between the constituent particles leads to a variety of interesting phenomena, are quantum magnets: these are quantum many-body systems, in which the interplay between immobile spin degrees of freedom (typically the spins of the electrons in a material, or pseudo-spin degrees of freedom, e.g., in cold gases experiments) leads to unconventional quantum phases. In this chapter, I will present various aspects of the phenomenology of quantum magnetism, the unconventional states of matter that can be realized, and how to characterize them. The topic is too vast to cover all essential aspects in a single book chapter. Therefore, most of the sections are summaries of (review) articles, text books, or from the introduction of PhD theses on related topics, and are intended as a starting point for further reading.

To start with, I will first put quantum magnetism in the context of strongly correlated quantum many-body systems and discuss typical models and examples. In order to understand what kind of states of matter can be obtained in such correlated systems, the basic notions of spontaneous symmetry breaking, order parameters, long-range order and topological order are discussed. Important experimental tools are spectroscopic measurements (e.g. inelastic neutron scattering), which will lead us to the notion of dynamical structure factors. At the end, I will give a glimpse onto recent developments, where the dynamical properties are further studied by going out of equilibrium and directly measuring the time evolution of the observables – typically, the demagnetization dynamics on very short time scales, or the time evolution of order parameters, which show that one can realize (transient) magnetically ordered states when exciting certain materials with a laser pulse.

1.1 Quantum many-body systems

Quantum many-body effects come into play in certain materials and in systems of ultracold atomic and molecular gases on optical lattices [1]. Prominent examples for strongly correlated materials are high-temperature superconductors [2, 3] and Mott insulators [4]. Using ultracold atoms, a breakthrough experiment was the realization of a Mott-insulating state of ultracold bosons in 2002 [5]. These systems are described by microscopic quantum mechanical models of interacting particles on various lattice geometries. Interesting effects arise due to competing interactions or geometrical frustration, which typically does not allow for the realization of a simple ground state that satisfies all bonds equally well. Examples for such frustrated geometries are shown in Fig. 1.

An interesting class of strongly correlated materials, in which such competing interactions lead to novel and interesting states of matter, are (frustrated) quantum magnets [11–14]. These systems can be described as networks of interacting quantum mechanical spins on a lattice, and

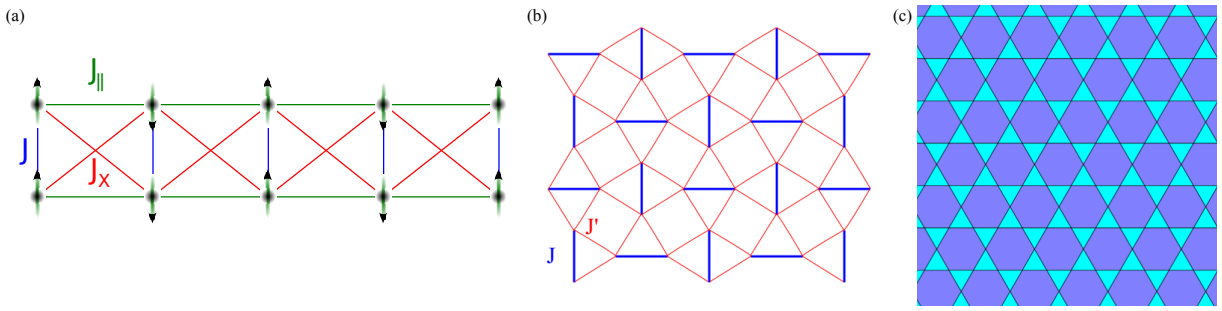

Fig. 1: *Examples of frustrated lattice geometries realized in quantum magnetic materials. (a) Frustrated ladder, realized in various materials, e.g., $TlCuCl_3$ [6, 7]. (b) Shastry-Sutherland lattice, a network of orthogonal dimers. This geometry is realized, e.g., in $SrCu_2(BO_3)_2$ [8, 9]. (c) Kagome lattice of corner-sharing triangles. This system is realized, e.g., in Herbertsmithite $\left(ZnCu_3(OH)_6Cl_2\right)$ [10].*

the underlying microscopic model typically is a variant of the Heisenberg Hamiltonian,

$$\mathcal{H}_{\text{Heisenberg}} = \sum_{\langle i,j \rangle} J_{i,j}\, \vec{S}_i \cdot \vec{S}_j, \tag{1}$$

where the operator $\vec{S}_i \equiv \begin{pmatrix} S_i^x \\ S_i^y \\ S_i^z \end{pmatrix}$ describes a localized spin on the lattice site i, and $S_i^{x,y,z}$ are the usual spin operators. In principle, any magnitude of $S \equiv \left|\vec{S}\right|$ can be considered, but the most appealing effects due to the quantum nature of the spins are expected for small values of S, e.g., $S = 1/2$ or $S = 1$. Many quantum magnetic materials can be described using the Heisenberg model or one of its variants and a rich bouquet of interesting phenomena is found, which are often revealed in the presence of an external magnetic field. For example, an *a priori* not necessarily expected realization of an unconventional phase of matter in a magnetic material is the Bose-Einstein-condensation (BEC) of triplet excitations (see, e.g. [7, 15–17]). Since these excitations are of bosonic nature, at temperatures low enough, they can form a BEC, so that quantum magnets can host this peculiar state of matter, which was first realized in the lab in experiments with ultracold gases [1]. Other quantum states of matter realized in these materials are Mott-insulators on magnetization plateaux [8, 9, 18, 19], and the proposed spin-equivalent [20–25] of a supersolid phase [26, 27], which is characterized by the simultaneous spontaneous breaking of the translational symmetry of the underlying lattice and of a U(1) symmetry associated to the formation of a superfluid. These effects most prominently appear at low temperatures, at which quantum fluctuations dominate over thermodynamic fluctuations, and which drive quantum phase transitions [28, 29].

Quantum states of matter are either described by local order parameters, which are due to the spontaneous breaking of a symmetry of the Hamiltonian, or by topological properties. The Heisenberg model (1) contains a scalar product of two vectors $\vec{S}$ and is, hence, invariant against rotation in spin-space, i.e., it possesses a SU(2) symmetry. An important question for quantum magnets is, therefore, if and how this symmetry (and possibly other symmetries) of the system

is broken spontaneously. The most prominent way of breaking the SU(2) symmetry is by realizing a finite magnetization, i.e., the expectation value of the spin in (at least) one direction in space gets finite, e.g. $\langle S_i^z \rangle \neq 0$. Since the magnetization is a vector and points in a specific direction, a finite magnetization implies a broken time-reversal symmetry. The question arises, if other (for quantum magnets less obvious) types of order can be realized. A prominent example is realized for $S > 1/2$: In this case, the SU(2) symmetry of the Heisenberg Hamiltonian can spontaneously be broken *without* resulting finite local magnetizations. This leads to a rather unconventional ordered phase which in an experimental investigation would appear disordered, if only (local or total) magnetizations are measured. Indeed, the resulting type of order is reminiscent of liquid crystals, which realize nematic states with a broken spin-rotational symmetry but unbroken time reversal symmetry [30, 13]. Correspondingly, such states are called *spin-nematic states* and have been explored in a large number of theoretical approaches (a nice summary is sketched in the introduction of [31] and references therein). Recently, the observation of such a spin-nematic state in an iridate material was reported [32].

One particular playground for quantum magnetic systems are phases in which, despite the presence of strong correlations in the system, *no* long-range order is induced at zero temperature. These phases are called *spin liquids* and can be pictured as a superposition of many spins which simultaneously point in different directions. They show exotic behavior and possess a number of interesting properties such as excitations with fractional quantum numbers [33]. There exists a vast literature on this topic, for an introduction see [34]. The search for realizations of this type of unconventional states is motivating a lot of ongoing research. For example, numerical methods based on tensor network states (TNS), in particular matrix product state (MPS) approaches, have provided evidence for the existence of such a spin liquid phase in the kagome lattice [35–37]. This highly frustrated geometry is depicted in Fig. 1(c) and has been identified, e.g., in the natural mineral Herbertsmithite [10, 38].

While the interacting spins in the Heisenberg model remain localized, in many materials the electrons are itinerant. This is addressed, e.g., by the Hubbard model [39–42], which is one of the simplest models taking into account the effects of spin and of electron motion. Both, the fermionic version[1]

$$\mathcal{H}_{\text{Hubbard}}^{\text{Fermions}} = -t \sum_{\langle i,j \rangle, \sigma} \left(c_{i,\sigma}^\dagger c_{j,\sigma} + h.c. \right) + U \sum_i n_{i,\uparrow} n_{i,\downarrow} \tag{2}$$

as well as the bosonic variant [28, 43]

$$\mathcal{H}_{\text{Hubbard}}^{\text{Bosons}} = -J \sum_{\langle i,j \rangle} \left(b_i^\dagger b_j + h.c. \right) + \frac{U}{2} \sum_i n_i (n_i - 1) \tag{3}$$

are relevant for the description of strongly correlated materials or for systems of ultracold atoms on optical lattices, respectively. Using degenerate perturbation theory, the Hamiltonian (2) in

[1] Standard notation for the operators is used, i.e., $c_{i,\sigma}^{(\dagger)}$ represents a fermionic annihilation (creation) operator, $b_i^{(\dagger)}$ the corresponding bosonic one, and $n_{i,\sigma} = c_{i,\sigma}^\dagger c_{i,\sigma}$ or $n_i = b_i^\dagger b_i$ the densities in the fermionic or bosonic case, respectively. In the case of the t-J-model, the operators $f_{i,\sigma}^{(\dagger)}$ are fermionic ones, but act on a restricted Hilbert space in which double occupancies are forbidden.

the strong coupling limit $U/t \gg 1$ can be mapped to the Heisenberg model (1) [44] with antiferromagnetic (AFM) interactions ($J > 0$ in the convention used in Eq. (1)). In this way, studying Hubbard systems allows one to study quantum magnetism. Due to the difficulties to treat in particular the fermionic variant of the Hubbard model beyond 1D using analytical or numerical approaches, experiments on optical lattices have got the particular motivation to *emulate* the behavior of this microscopic model [45], so that its phase diagram can be investigated in such experiments. This is in the spirit of Feynman's proposal from the early 1980s[2] to use some well controlled quantum systems to simulate other ones, eventually leading to the development of a quantum computer [47–49]. In this way, for $U/t \gg 1$ quantum magnetism can be studied, and the realization of a controlled quantum simulator for quantum magnetism in cold gases experiments is a central topic of ongoing research. Interestingly, there are further proposals for how to realize Heisenberg-type models in cold gases experiments based on ultracold polar molecules, e.g., where internal degrees of freedom of the molecules can be used as pseudo-spin degrees of freedom (see, e.g., [50, 51]).

One interesting hybrid of itinerant electrons and Heisenberg exchange is the so-called t-J model

$$\mathcal{H}_{\mathrm{tJ}} = -t \sum_{\langle i,j\rangle,\,\sigma} \left(f^{\dagger}_{i,\sigma} f_{j,\sigma} + h.c. \right) + J \sum_{\langle i,j\rangle} \left(\vec{S}_i \cdot \vec{S}_j - \frac{1}{4}\, n_i n_j \right), \tag{4}$$

which, similarly to the Heisenberg model, can be obtained from the fermionic Hubbard model in degenerate perturbation theory in the limit $U/t \to \infty$ [44] and by excluding double occupancies on the lattice sites. One possible way to look at the t-J model is to imagine a lattice with initially one spin per site, in which the spins interact via Heisenberg exchange, but then *dope* it (i.e. remove more and more of the electrons). In this way, empty lattice sites are formed, and the spins can 'hop around' in addition to experiencing the spin exchange interaction. Since in cuprates high-temperature superconductivity is obtained by doping an AFM, there are proposals to understand high-temperature superconductors based on spin fluctuations, and the t-J model (which describes such a doped quantum magnet) is considered a minimal model [3, 52].

The fermionic and spin systems considered so far have a SU(2) symmetry and are invariant under the corresponding transformations. From the theoretical side, it is tempting to enhance this symmetry from SU(2) to SU(N). This has attracted considerable theoretical attention. There is a long history of studies of SU(N) spin systems (see, e.g., [53–55]) since they become analytically tractable in the large-N limit, and rich phase diagrams have been identified. In 1D, the aforementioned spin-nematic phases have been predicted as well as generalizations of the so-called AKLT state, which is an archetypical example for topological phases which are briefly revisited in Sec. 2.2.

Since no exact SU(N) models have been identified in nature, these efforts were broadly considered a theoretical playground. However, it has been proposed that systems with such a high symmetry (up to $N = 10$) can be realized in quantum simulators with ultracold alkaline earth

[2] The possibility to exploit quantum speed up was actually first envisaged by Y.I. Manin [46] in a radio interview with Radio Moscow in 1980; it is unclear to me whether Feynman was aware of this. In any case, he seems to be the first one to promote these ideas in the public in the western hemisphere and pursue them.

atoms [56]. More specifically, these experiments can realize SU(N) symmetric generalizations of fermionic Hubbard models

$$\mathcal{H}^{\mathrm{SU}(N)}_{\mathrm{Hubbard}} = -t \sum_{\langle i,j\rangle} \sum_{\alpha=1}^{N} \left(f^{\dagger}_{\alpha,i} f_{\alpha,j} + h.c. \right) + \frac{U}{2} \sum_{i,\alpha\neq\alpha'} f^{\dagger}_{\alpha,i} f^{\dagger}_{\alpha',i} f_{\alpha',i} f_{\alpha,i} \,. \tag{5}$$

Here, $f^{(\dagger)}_{\alpha,i}$ is a fermionic annihilation (creation) operator for a particle with flavor α on lattice site i. Similarly to the SU(2) case, in the limit $U/t \to \infty$ an effective SU(N) symmetric Heisenberg model can be derived

$$\mathcal{H}^{\mathrm{SU}(N)}_{\mathrm{Heisenberg}} = \frac{2t^2}{U} \sum_{\langle i,j\rangle} \sum_{\alpha,\beta} S^{\beta}_{\alpha}(i) S^{\alpha}_{\beta}(j) \,, \tag{6}$$

with the spin operators $S^{\beta}_{\alpha}(i) = f^{\dagger}_{\alpha,i} f_{\beta,i}$. This opens the door to studying SU(N) quantum magnetism. Having this and the specific experimental implementation in mind, exotic new phases have been predicted. An example, which has intrigued researchers, is the possibility to realize chiral spin liquids [57, 58] in such systems [59]. These are spin liquids with certain topological properties, which can be of relevance for the realization of topological quantum computers [60, 33].

These findings underline the recent focus of research on the uncovering of new and unconventional behavior in microscopic models and their possible experimental detection. Since most of the models are non integrable,[3] numerical methods play an important role in the investigation of quantum magnetism. A very powerful approach is using *quantum Monte Carlo* techniques [61], which, in principle, can be applied to arbitrary situations. However, fermionic and AFM frustrated spin systems are affected by what is known as 'the sign problem', which leads to negative probabilities in the course of the Monte Carlo sampling, and which makes it essentially impossible to control the calculations for many interesting situations. Therefore, other, wavefunction based approaches have been developed. For quasi-1D systems (i.e., chains and ladder geometries), efficient approaches are *tensor network methods*, in particular *matrix product state* methods (MPS) and one of the realizations in terms of the density matrix renormalization group method (DMRG) [62], which are explained in detail in various review articles (e.g. [63]). This method has been applied very successfully for the investigation of phase diagrams and of quantum critical behavior of a multitude of (quasi-)1D systems (see the website [64] for a collection of the publications relying on this method). However, for 2D systems, the area law of entanglement growth [65, 66] is a major obstacle for an efficient treatment of the microscopic models of interest. Since also other numerical methods are limited, it is an ongoing challenge to develop numerical and analytical approaches for 2D quantum magnetic systems.

[3] An important exception is the Heisenberg model and its generalization to the XXZ-model by allowing the spin-exchange in the z-direction to have a different strength than in the x-y-plane in 1D, which is integrable using the Bethe ansatz [42].

1.2 Basic properties of $S = 1/2$ quantum magnets, magnetization curves

In order to get a better intuition for the behavior of quantum magnets, it is useful to start with a small number of quantum spins, which can serve as building blocks to understand the large interacting networks of spins realized in quantum magnets. To do so, let us consider a $S = 1/2$ Heisenberg model with interactions between nearest neighboring spins in a magnetic field $\vec{B}$,

$$\mathcal{H} = J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j - \vec{B} \sum_j \vec{S}_j \,. \tag{7}$$

To simplify the discussion, we assume the magnetic field $\vec{B}$ to point in z-direction.

1. One single spin-1/2:
   The Hamiltonian is simply $\mathcal{H}{=}-BS_i^z$, i.e., in its ground state the spin can take one of the two possible configurations $|\uparrow\rangle$, $|\downarrow\rangle$, depending on the direction the magnetic field is pointing at.
   Note: for $B = 0$, any superposition of both states is a possible ground state, $|\psi\rangle = \alpha\,|\uparrow\rangle + \beta\,|\downarrow\rangle$ with $\alpha^2 + \beta^2 = 1$, and for $\alpha = \beta$ a so-called "cat state" is realized.[4] This can be relevant for quantum computation, where you can associate the two spin states to the two possible internal states of a *qubit* (e.g., $|\uparrow\rangle \equiv |1\rangle\,,\ |\downarrow\rangle \equiv |0\rangle$).

2. Two spin-1/2 objects interacting via Heisenberg exchange (spin-1/2 dimer):
   The Hamiltonian now is

   $$\mathcal{H} = J\vec{S}_1 \cdot \vec{S}_2 - B\,(S_1^z{+}S_2^z) = J\Big(\frac{1}{2}\big(S_1^+S_2^-{+}S_1^-S_2^+\big) + S_1^zS_2^z\Big) - B\big(S_1^z{+}S_2^z\big)\,, \tag{8}$$

   with ladder operators $S^\pm = S^x \pm iS^y$.
   Let us first consider the case $B = 0$. In this case, the Hamiltonian has the full SU(2) symmetry, and we can rewrite

   $$\vec{S}_{total} = \vec{S}_1{+}\vec{S}_2 \ \Rightarrow\ \vec{S}_1 \cdot \vec{S}_2 = \frac{1}{2}\left(\big(\vec{S}_{total}\big)^2 - \big(\vec{S}_1\big)^2 - \big(\vec{S}_2\big)^2\right)\,.$$

   Realizing that $\vec{S}^2 = S(S{+}1)$, we obtain for the Hamiltonian (8) for $S = 1/2$ at $B = 0$

   $$\mathcal{H} = \frac{J}{2}\Big(\big(\vec{S}_{total}\big)^2 - \frac{3}{2}\Big).$$

   Since for $S = 1/2$ the only possible values for $\vec{S}_{total}$ are 0 or 1, respectively, we see immediately that the Hamiltonian has only two eigenvalues

   $$E_{S_{total}=0} = -3J/4 \quad \text{and} \qquad E_{S_{total}=1} = J/4\,.$$

   Depending on the sign of J, either of the two values is the ground state. Due to the degeneracy (see below) the state with $S_{total}{=}0$ is called *singlet state* (it is not degenerate), and the other one is threefold degenerate and accordingly the eigenstates are called *triplet states*.

[4] Named after the famous gedankenexperiment with "Schrödinger's cat".

We now turn to the Hamiltonian matrix, i.e., we need to introduce a suitable many-body basis, which can be obtained by the tensor product of single-spin basis states, leading to the basis states $\{ |\downarrow\downarrow\rangle , |\downarrow\uparrow\rangle , |\uparrow\downarrow\rangle , |\uparrow\uparrow\rangle \}$. Note that the number of basis states compared to one spin-$1/2$ particle has *doubled*. This is at the heart of why it is so complicated to numerically treat large quantum many-body systems: here, the dimension d of the basis grows exponentially with the number N of spins in the system, $d = 2^N$, so that only small systems can be treated exactly [61].
For the dimer, the Hamiltonian in this basis is represented by the matrix

$$\mathbf{H} = \begin{pmatrix} \frac{J}{4}+B & 0 & 0 & 0 \\ 0 & -\frac{J}{4} & \frac{J}{2} & 0 \\ 0 & \frac{J}{2} & -\frac{J}{4} & 0 \\ 0 & 0 & 0 & \frac{J}{4}-B \end{pmatrix} . \tag{9}$$

Note the *block structure* on the diagonal of the matrix. This is due to symmetries and conserved quantities (here: conservation of the z-component of the total spin of the system) and follows from Schur's Lemma [67]. It can be exploited to speed up the diagonalization of the matrix, since one needs to explicitly diagonalize only the smaller 'blocks' of the matrix, i.e., for Hamiltonian (9) we need to diagonalize only a 2×2 matrix. This leads to the following eigenstates and eigenvalues:

- Singlet-state $|s\rangle = \big(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \big)/\sqrt{2}$, with energy $E_s = -3J/4$. This is the state with $S_{total} = 0$, which we encountered above, and also the z-component $S^z_{\text{total}} \equiv \big\langle S^z_1 + S^z_2 \big\rangle = 0$, so that the magnetic field B does not contribute to the energy.
  Note: This state is a *maximally entangled* state and is *antisymmetric* when swapping the position of the two spins. Indeed, it is one of the *Bell states* and can be useful for quantum information aspects.

- Triplet-states:

$$\begin{array}{lll} |t_1\rangle = |\uparrow\uparrow\rangle & |t_0\rangle = \big(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \big)/\sqrt{2} & |t_{-1}\rangle = |\downarrow\downarrow\rangle \\ E_{t_1} = J/4 - B & E_{t_0} = J/4 & E_{t_{-1}} = J/4 + B \end{array}$$

  For all three states we have $S_{\text{total}} = 1$, but $S^z_{\text{total}} = +1$, 0, or -1, respectively. We see that at $B = 0$ the three states are degenerate, but that turning on a magnetic field removes this degeneracy: depending on the sign of B, the energy of one of the triplet states $|t_1\rangle$ or $|t_{-1}\rangle$ will grow linearly with B, the other one decreases linearly with B; the energy of $|t_0\rangle$ remains unchanged. Using this, we obtain our first magnetization curve: for $J > 0$ (AFM) the singlet state is the lowest energy state for $|B| < J$, but then one of the triplet states takes over, so that we have a jump in the total magnetization from $M = 0$ to $M = 1$ at this 'critical field strength'.

  Note: The states $|t_1\rangle$ and $|t_{-1}\rangle$ are product states with zero entanglement, but $|t_0\rangle$ is maximally entangled, as is also the singlet state (it is another Bell state); however, all three triplet states are symmetric when swapping the positions of the spins.

The physics of such spin-$1/2$ dimers is the building block for various quantum magnets. A prominent example is the Shastry-Sutherland lattice depicted in Fig. 1(b), and which can be seen as a system of orthogonal dimers, where the coupling between the dimers can vary. Other systems are, e.g., spin ladders (see Fig. 1(a)), in which the interaction on the rungs is stronger than on the legs. For weak inter-dimer coupling, the physics of such quantum magnetic systems is determined by the properties of the singlet and triplet states discussed here. The inter-dimer coupling will lead to a 'dressing' of the triplet states (resulting in so-called 'triplons'), which then form the building block for the system's behavior. When such a system with AFM interactions on the dimers is put in a magnetic field, the ground state at $B = 0$ is determined by the singlets, but the magnetization curve will mainly be determined by the magnetization of the individual dimers: the dimers, which at $B = 0$ are in a singlet state, are 'populated' by the triplets (or triplons) upon increasing B, leading to particular behavior of the magnetization curve. For example, one can derive effective models $\big($e.g. using perturbative unitary transformations, PCUTs; see, e.g., [68]$\big)$, in which the triplons interact via long-range interactions, and which can form Wigner-type crystals. If this happens, a gap opens, and a magnetization plateau is stabilized, which hosts a Mott insulator, which is formed by the crystal of triplons. Other examples are the aforementioned possible Bose-Einstein condensation of these triplet or triplon excitations at finite B.

3. Three spin-$1/2$ objects interacting via Heisenberg exchange:
   As before, at $B = 0$, we can rewrite using $\vec{S}_{total}$ and obtain
   $$\mathcal{H} = \frac{J}{2}\Big(\big(\vec{S}_{\text{total}}\big)^2 - \frac{9}{4}\Big) \quad \text{for } S = 1/2\,.$$
   We realize that, as in the system with 2 spins, the ground state for the ferromagnetic (FM) case $J < 0$ is obtained by maximizing $\vec{S}_{\text{total}}$, while for the AFM case $J > 0$, $\vec{S}_{\text{total}}$ needs to be minimized. For $S = 1/2$, the maximal value of $|\vec{S}_{\text{total}}| = 3/2$, and all spins point in the same direction. All the interactions on the bonds are satisfied. However, in the AFM case, the ground state energy is minimal for the smallest possible value $|\vec{S}_{\text{total}}| = 1/2$. We realize the following two aspects:
   i) There are in total *6 configurations*, which all result in $|\vec{S}_{\text{total}}| = 1/2$, i.e., the ground state of the AFM case will be a superposition of the corresponding 6 states: it is *highly* degenerate, since in total we have only $2^3 = 8$ basis states, and hence the largest part of the Hilbert space contributes to the ground state manifold!
   ii) There is no way to simultaneously satisfy all the three interaction terms (i.e., minimize the energy of each of the three bonds): the system is the simplest example for a *geometrically frustrated* quantum magnet. One effect of such geometrical frustration is that the spins try to find a new configuration, in which none of the bonds is fully satisfied, but which minimizes the total energy. As such, qualitatively new behavior can be realized when comparing to non-frustrated geometries. One example is the classical Heisenberg model on a triangle, where the three spins align in the plane and point outwards, forming a 120° angle between each other.

There are many quantum magnetic materials with such AFM triangles as building blocks, see Fig. 1 for example geometries. Particularly interesting situations are obtained when a lattice consists of corner-sharing triangles, such as the kagome lattice depicted in Fig. 1(c). Here, the degree of frustration in the extended lattice is very high and determines the ground state physics. As we have seen for a single triangle, ground states of frustrated AFM are typically very highly degenerate. Due to this degeneracy, the spins fluctuate strongly, so that under circumstances they cannot realize long-range order. This leads to a peculiar situation: the ground state and the excited states are governed by the interactions between the spins, but, due to the strong fluctuations, any attempt to stabilize long-range order is suppressed. Such a state is called a *spin liquid*. Such spin liquid states can also host topological phases of matter (which do not rely on the existence of a finite local order parameter, but are described by global quantities), and due to their rich and unconventional behavior have been a focus of intense studies.

We could, of course, go on, and discuss larger and larger building blocks. Noteworthy are lattices, in which the building blocks are tetrahedrons; for ferromagnetic Ising-type interactions $\mathcal{H}^{\text{Ising}} = J\sum_{\langle i,j\rangle} S_i^z S_j^z$, again a highly frustrated geometry can be realized, e.g., on so-called pyrochlore lattices. The spins align according to so-called 'spin-ice rules' (two spins point into the tetraeder, two point outwards), and due to the high frustration, unconventional states can be realized. Indeed, such systems can even host excitations, which can be described as magnetic monopoles, which in vacuum cannot exist, but are realized and measured in these systems [69, 70].
The generic question is how to deal with a large number of spins on an arbitrary graph. Since in most cases there is no analytical solution, but we are dealing with finite-dimensional Hilbert spaces, we can use the matrix representation of the Hamiltonian in an appropriately chosen basis and diagonalize this on a computer. We refer the reader to, e.g., Refs. [61, 71].

1.2.1 Magnetization curves

An important quantity to study in quantum magnets is the dependence of the magnetization on the applied magnetic field $\vec{B}$, which typically is included in the Hamiltonian as a Zeeman-type term (see Eq. (7)) and which, for the sake of simplicity, we will assume is pointing in z-direction. In this case, the original SU(2) symmetry of the Heisenberg model is broken down to U(1), which can be exploited for investigating the system. Note that, however, the most general form of the Zeeman term is $\sim \vec{B}\mathbf{g}\vec{S}$, with the so-called '$g$-tensor' $\mathbf{g}$, which captures a possibly anisotropic response of the material to the applied magnetic field. If a material has a non-trivial g-tensor, also the remaining U(1) symmetry is broken, and the magnetization will not point in parallel direction to $\vec{B}$. This can lead to further interesting behavior as, e.g., a torque on the sample. Here, however, for the sake of simplicity we assume a simple g-tensor and hence a response only in parallel direction to $\vec{B}$, so that the U(1) symmetry prevails. Then, it suffices to treat the magnetization $M=\langle S^z_{total}\rangle$, which can have nontrivial behavior. For example, if there is an excitation gap, then increasing the magnetic field will not change the value of M, leading to

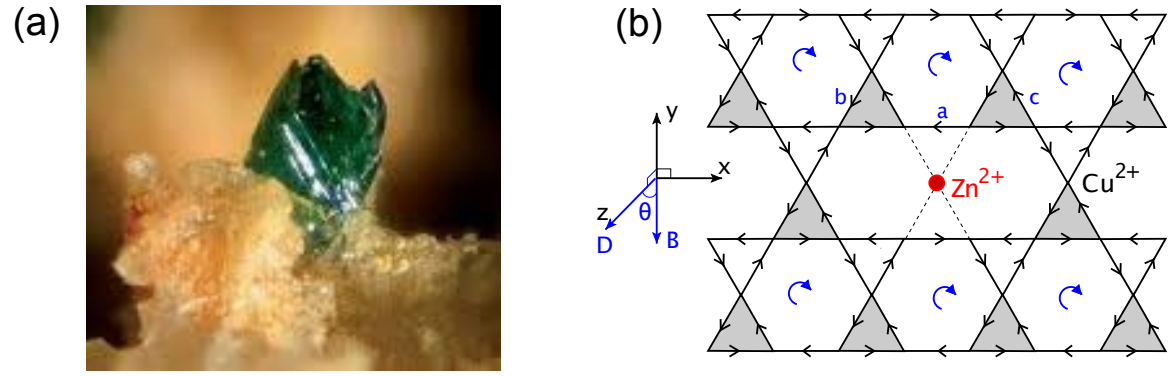

Fig. 2: *(a) The natural mineral Herbertsmithite is a realization of a Heisenberg spin-$^1/_2$ system on the kagome lattice [10]. However, the material contains $\sim 5\%$ nonmagnetic impurities and anisotropic DM interactions due to spin-orbit coupling. (b) The lattice with one impurity and the orientation of the DM-vectors used in [74].*

a *magnetization plateau*, as already illustrated further above in the context of the spin-$^1/_2$ dimer. These plateaux can host interesting Mott phases (e.g., crystals of triplets). In the vicinity of these magnetization plateaux, it is interesting to study how the plateau 'melts', and the resulting phase can host unconventional states, like the BEC of triplets, or even supersolid phases.
For systems with U(1) symmetry in a magnetic field, the total value of S^z_{total} is conserved, so that one does not need to compute explicitly $M(B)$ via expectation values. Instead, at zero temperature,[5] one computes the ground state energies $E_0(S^z_{total})$ in all possible sectors of S^z_{total} and obtains the Magnetization via a Legendre-transform:

$$M(B) = \left\langle S^z_{total} \right\rangle \Big|_{\left[E_0(S^z_{total}, B=0) - B \cdot S^z_{total}\right] = \min} .$$

Therefore, it is important to have methods to efficiently compute the ground state energy for systems as large as possible in order to be as close as possible to the thermodynamic limit. Two such approaches are the aforementioned exact diagonalizations (ED) and tensor network states, such as the MPS or the Projected Entangled Pair States (PEPS) [72]. Both methods also exist in a version, which works directly in the thermodynamic limit as iMPS and iPEPS, respectively. In particular the iPEPS has been successful in investigating magnetic properties of dimer-based quantum magnets such as the Shastry-Sutherland lattice, see, e.g. [73].

1.3 Effect of spin-orbit coupling at high magnetic fields

The models mentioned so far are often minimal models. However, in real materials additional effects like the anisotropic g-tensor mentioned above, or other anisotropies due to spin-orbit coupling (SOC) are present. In a magnetic field, SOC can alter the physics of the system significantly [75], but is often neglected, since its magnitude typically is only a few percent of the magnitude of the Heisenberg exchange in the system. However, since its effect can be

[5] Remember that we are interested in low temperature properties, since here quantum fluctuations will have a stronger effect than thermal ones.

important, in order to obtain a more realistic description, it is necessary to consider SOC by treating additional anisotropic interactions, e.g., of Dzyaloshinskii-Moriya (DM) type [76, 77],

$$\mathcal{H}_{\mathrm{DM}} = \sum_{\langle i,j \rangle} \vec{D}_{i,j} \cdot \left(\vec{S}_i \times \vec{S}_j \right). \tag{10}$$

This term arises in the strong coupling limit of the fermionic Hubbard model (2) when taking into account spin orbit coupling $\sim \vec{L} \cdot \vec{S}$. Note that, in contrast to the Heisenberg term (1), the DM interaction is antisymmetric upon exchange of the spins and breaks the SU(2) symmetry. On dimers, this leads to a mixing of the singlet and the triplet sectors and can so lead to new interesting effects. An example for DM interactions in a kagome system is depicted in Fig. 2.

2 Unconventional phases and quantum critical behavior

In the previous section, we have seen the most important microscopic models for quantum magnetism (in particular the Heisenberg model (1)), and related models of itinerant fermions or bosons. Most of these models are inspired by quantum magnetic materials. One such material, which hosts unconventional behavior in magnetic fields, is $SrCu_2(BO_3)_2$, which is a very good realization of the AFM Heisenberg Hamiltonian on the Shastry-Sutherland lattice [78] depicted in Fig. 1(b) [8, 9], with additional DM interactions. Using a combination of various numerical methods, the magnetization curve up to 118 T has been compared to experimental data [79], and interesting Mott insulators on magnetization plateaux have been found (see Fig. 3), e.g., one which is formed by bound states of triplons [73]. Another example for a quantum magnetic material, which hosts an unconventional phase, is the ground state of the kagome lattice in Hebertsmithite, which realizes the Heisenberg Hamiltonian with additional DM interactions on this lattice. However, a large number of non-magnetic impurities is present in this material, so that additional effects close to the impurity sites come into play (see Fig. 2).

Many studies are performed on a large variety of further quantum magnetic materials. However, also beyond their possible realization in a material it is interesting *per se* to formulate models with competing interactions based on mathematical insights and to investigate their properties. An example we already encountered in Sec. 1.1 is to enhance the SU(2) symmetry to SU(N). Another model that can be introduced in this line of thinking is the $S = 1$ Heisenberg chain with additional biquadratic interactions in a magnetic field,

$$\mathcal{H} = \sum_i \left(\cos(\theta)\, \vec{S}_i \cdot \vec{S}_{i+1} + \sin(\theta) \left(\vec{S}_i \cdot \vec{S}_{i+1} \right)^2 \right) - B \sum_i S_i^z \,, \tag{11}$$

the so called *bilinear biquadratic* Hamiltonian (BLBQ). Here, at $B = 0$, the simple Heisenberg model of Eq. (1) has been extended by a term in such a way that the resulting Hamiltonian still possesses the SU(2) symmetry of the Heisenberg model,[6] but the Hamiltonian now has two competing interaction terms. That the two terms compete with each other can be seen by considering a spin-1 dimer: while for the bilinear term (the Heisenberg term) the ground state

[6] The biquadratic term itself has, actually, SU(3) symmetry.

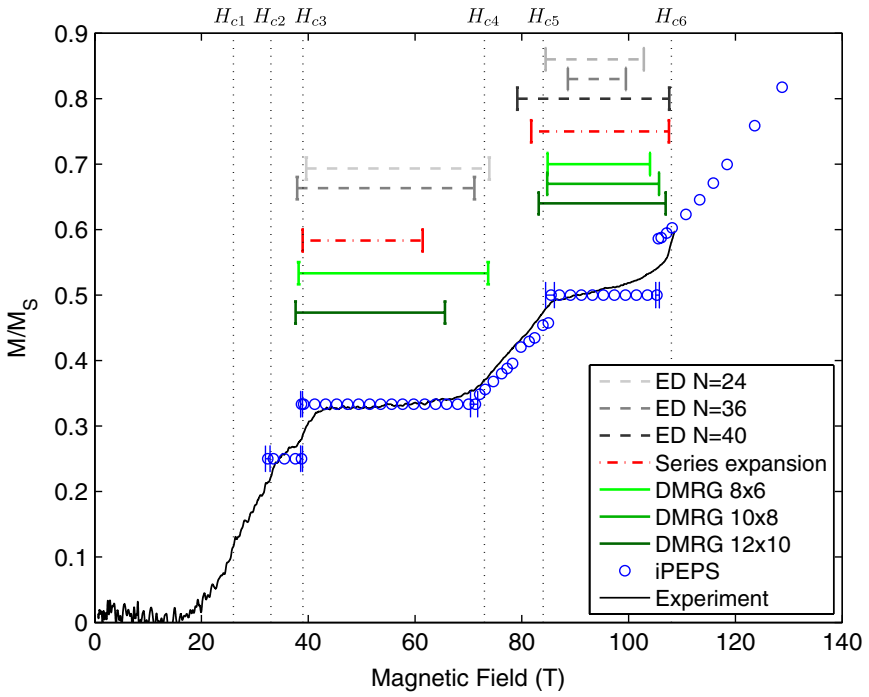

Fig. 3: *Comparison of the experimental data for the magnetization curve of $SrCu_2(BO_3)_2$ to theoretical results obtained with different approaches (Fig. taken from [79]).*

is either a singlet or a triplet, for the biquadratic term it can be a *quintet* state with $S_{total} = 2$. Going to an extended lattice, one can therefore expect that at zero and at finite B different types of ground states will compete with each other, depending on the strength and the sign of the respective bilinear or biquadratic term. As discussed in [80], the resulting phase diagram of (11) is, indeed, rich. Interestingly, one of the phases realizes spin-nematic quasi long range order (QLRO, see further below). The question arises, how to systematically characterize the different types of order that can emerge in such systems.

2.1 Equilibrium: order parameters from symmetry considerations

Here we give a summary of important aspects concerning spontaneous symmetry breaking. For further reading, I suggest, e.g., the excellent lecture notes 'An introduction to spontaneous symmetry breaking' by A.J. Beekman *et al.* [81].

In Landau's theory of phase transitions, spontaneous symmetry breaking (SSB) leads to a finite local order parameter. In such a scenario, the state of the system[7] is not symmetric under a symmetry transform U, which leaves the Hamiltonian $\mathcal{H}$ invariant $\big($i.e. $[\mathcal{H}, U] = 0\big)$; the symmetry of the state is 'lower' than the symmetry of the Hamiltonian. Since symmetries are mathematically described by groups, the state can then still be symmetric under a transform corresponding to a subgroup H of the original symmetry group G of the Hamiltonian, if it preserves any symmetry at all.

[7] Typically, we are interested in the ground state $|\psi_0\rangle$ since many of the phenomena discussed here are realized at low temperatures, but the considerations also hold for the thermal equilibrium state.

Is such a symmetry broken state unique? To answer this, consider the following: $[\mathcal{H}, U] = 0$, so we know that an eigenstate $|\psi\rangle$ of $\mathcal{H}$ and $U|\psi\rangle$ must have the same energies, since $|\psi\rangle$ is a simultaneous eigenstate of $\mathcal{H}$ and U. However, $|\psi\rangle \neq U|\psi\rangle$ if the symmetry is spontaneously broken, i.e., in the case of SSB, multiple related states exist, which all share the same energy. Indeed, there exists a whole set of distinct symmetry-broken states with the same energy, which can be obtained by performing all possible symmetry transforms U on the symmetry broken state $|\psi\rangle$. These states are, hence, all related to each other by the symmetry G. This allows us to define the order parameter $\mathcal{O}$: it is the operator, whose eigenstates are the inequivalent states in the set of symmetry related states, and whose eigenvalues are different *and* non-zero for each of these states. $\mathcal{O}$ is constructed in such a way, that it has eigenvalue zero for states, which are symmetric under the transform U. Note however, that due to the so-called orthogonality catastrophe, for a finite system, $\langle\psi|\mathcal{O}|\psi\rangle = 0$ [81]. We will come back to this later, when we ask how to investigate order parameters in practice.

How to think about order parameters? Can we find a way to construct them? This is not straightforward to answer, and we have to dive a little deeper into the mathematics of symmetry transformations. From group theory we learn that in the thermodynamic limit (we will come back to this further below), one can classify the symmetry broken states by the *cosets* gH, which are elements of the *quotient set* G/H, if G is the group of all symmetry transforms, $H \subset G$ is the subgroup of unbroken transformations, and $g \in G$. H is then also called the *residual symmetry group*. For example, let us have a closer look at the SU(2) symmetry of the AFM Heisenberg model. Consider a Néel state $|\uparrow\downarrow\uparrow\downarrow\ldots\rangle$, in which the symmetry is broken down to rotations around a single axis, e.g., in z-direction. Hence, $G =$SU(2), while the residual symmetry group is $H =$U(1). One finds for the quotient set $G/H =$SU(2)/U(1)$\simeq S^2$, which gives all the points on the surface of a sphere. The direction of the sublattice magnetization is then one of these points on the sphere; hence, without applying an external field, infinitely many directions are possible, and the symmetry broken state will pick one of these.

For continuous symmetries, we consider the generators Q of the group, which can be introduced by considering transformations $U(d\theta)$ by an infinitesimally small value of a parameter θ (e.g., for rotations θ is the rotation angle), so that one can write $U(d\theta) \approx 1 + i\, d\theta\, Q$. This allows one to write for arbitrary angles $U(\theta) = e^{i\theta Q}$, which is obtained by subsequently applying transforms $U(d\theta)$ until the desired value θ of the parameter is obtained [67]. Consider a broken-symmetry state $|\psi\rangle$. Then, generators Q, of which $|\psi\rangle$ is an eigenstate, are called *unbroken generators*, and conversely generators, which do not leave the state invariant, are called *broken*. The dimension of the quotient set G/H is then said to equal the number of broken generators.

It would now be useful to identify an operator, whose expectation value can be used to distinguish between the symmetry broken states, and which has zero expectation value in the symmetric state. For each of the sets of equivalent symmetry-broken states, it should have a unique non-zero expectation value. This leads us to the question of how to identify suitable order parameters.

Let $U = e^{i\alpha Q}$ be a symmetry transform such that $[\mathcal{H}, U] = 0$. Since $U|\psi\rangle \neq |\psi\rangle$ for a symmetry broken state, this also holds for the generator, $Q|\psi\rangle \neq |\psi\rangle$, so that we cannot simply consider

the expectation value $\langle\psi\,|Q|\,\psi\rangle$. Instead, one proceeds as follows:
A state $|\psi\rangle$ breaks this symmetry, if there exists any operator Φ such that

$$\langle\psi\,|[Q,\,\Phi]|\,\psi\rangle \neq 0\,. \tag{12}$$

If no such operator exists, $|\psi\rangle$ is symmetric under U. $\Phi(x)$ is called interpolating field,[8] and allows us to introduce the order parameter operator $\mathcal{O}(x)$ and its expectation value, which then is the local order parameter:

$$\mathcal{O}(x) = \big[Q,\,\Phi(x)\big] \quad \text{and} \qquad O(x) = \langle\psi\,|\mathcal{O}(x)|\,\psi\rangle\,.$$

Due to Eq. (12), $O(x)$ is automatically zero if $|\psi\rangle$ is a symmetric state, and finite otherwise, so that it, indeed, distinguishes symmetric from symmetry-breaking states. Note that $\Phi(x)$ and $\mathcal{O}(x)$ are not necessarily hermitian, but one can always construct an observable from this operator, e.g., $\mathcal{O}+\mathcal{O}^\dagger$ or $\mathcal{O}\mathcal{O}^\dagger$.
It is possible to always find an operator $\mathcal{O}$ such that $O(x)$ will be different for distinct broken-symmetry states and equal for states related by residual symmetry transforms, since Eq. (12) does not uniquely determine $\mathcal{O}$ and Φ (e.g, construct an alternative interpolating field by multiplying Φ by a constant, then the equation can still be fulfilled). In almost all cases, the physics of the symmetry-breaking system itself suggests a convenient choice for $\mathcal{O}$, which maps onto the quotient space G/H. Furthermore, it inherits the structure of the quotient space.
Let us consider a concrete example for quantum magnets and go back to the Heisenberg AFM. The Hamiltonian has SU(2) symmetry, which is broken down in the AFM state to U(1). Inequivalent broken-symmetry states correspond to AFM configurations with the sublattice magnetization pointing in different directions – all possibilities together constitute the points on the surface of a sphere, S^2, as discussed above, and which coincides with the quotient SU(2)/U(1)$\simeq S^2$. We now choose the pointer along the z-direction. Hence, the symmetry generators S^x and S^y are broken, but not S^z. How to introduce an interpolating field? We expect for the AFM state the spins to alternately point in the up- and down-direction, respectively, so that it is natural to introduce the *staggered magnetization* $N_i^a = (\pm 1)^i S_i^a$, with i the position on the lattice, and $a = x, y, z$. Can we use N_i^a as interpolating field? Let us consider the breaking of rotations generated by S^x. We see after a short calculation, that $\sum_{ij}[S_i^x,\,N_j^y] = i\sum_i N_i^z$. Similarly, choosing S^y and N_j^x also leads to $\sum_{ij}[S_i^y,\,N_j^x] = i\sum_i N_i^z$. Hence, it seems plausible that the choice $\mathcal{O} = N^z = \sum_i N_i^z$ gives a suitable order parameter operator. Its expectation value then is the expectation value for the staggered magnetization, which can be measured in experiments (e.g. by measuring spin structure factors) – and which also would be the natural choice for an AFM state, since there we expect the spins to alternate, as in the classical Néel state. Note, however, that the classical Néel state is *not* an eigenstate of the AFM Heisenberg model, but, in the thermodynamic limit, if SSB takes place, the expectation value of the staggered magnetization is finite, *like* in a Néel state. This brings us to the following considerations:
In general, $[\mathcal{H},\,\mathcal{O}] \neq 0$, which would imply that the symmetry broken states are not eigenstates of $\mathcal{H}$, contradicting our above statement. In particular, for numerical methods, which treat finite

[8] We work for the moment with continuous variables x and will go back to lattice positions later.

system sizes, this implies that the expectation value $\langle\psi_0|\mathcal{O}|\psi_0\rangle = 0$ for all system sizes, so the question arises, how to investigate for the order characterized by $\mathcal{O}$, a topic we will return to later. The solution to this puzzle lies in the necessity of taking the *thermodynamic limit* $N \to \infty$ and $V \to \infty$, with $N/V = const$. It turns out that in this limit $\big\langle[\mathcal{H},\, \mathcal{O}]\big\rangle{=}0$, and the symmetry-broken states become orthogonal to one another in this limit, as well as degenerate with the symmetric exact eigenstates of $\mathcal{H}$. If in this limit, the symmetry-broken states are eigenstates of $\mathcal{H}$. The thermodynamic limit is, hence, always different to any finite volume (irrespective of its size), and makes it a *singular* limit. Care needs to be taken, when computing quantities in this limit. Often, in order to have some finite expectation value, one applies a small field[9] h, which induces the order one wants to investigate, but which one needs to 'remove' again. Hence, when studying observables in the thermodynamic limit, two limits need to be taken, and since the thermodynamic limit is singular, the order cannot be exchanged. Hence, the order parameter can be obtained as

$$\langle\mathcal{O}\rangle = \lim_{h\to 0}\lim_{N\to\infty}\langle\psi_0(h,N)|\mathcal{O}|\psi_0(h,N)\rangle\ ,$$

with $|\psi_0(h,N)\rangle$ being the ground state for a system with N spins and when applying a small field h.

From these considerations, two questions are imminent: i) how to compute order parameters numerically, if one treats finite systems? ii) which order parameters can we realize in a quantum magnetic system?

We first turn to the latter. As we have seen above, this needs some careful thought. Since we are dealing with systems on lattices, one can consider the breaking of the lattice symmetries, like translational, rotational, or parity symmetry. Since this is not peculiar for spin systems (also itinerant electrons on such lattices can undergo the corresponding SSB), we focus here on the SSB associated to the spin degrees of freedom, and discuss the above mentioned example of spin-nematic order as an unconventional way to realize SSB in quantum magnets.

2.1.1 Spin-nematic order

We start by describing an $S = 1$ object by the three S^z eigenstates $|{\downarrow}\rangle$, $|0\rangle$, $|{\uparrow}\rangle$. From these, we can construct basis states (see also the Suppl. Material of [82] and the PhD thesis of T. Tóth (EPF Lausanne, 2011) [83])

$$|x\rangle = \frac{i}{\sqrt{2}}\big(|{\uparrow}\rangle - |{\downarrow}\rangle\big)\,,\quad |y\rangle = \frac{1}{\sqrt{2}}\big(|{\uparrow}\rangle + |{\downarrow}\rangle\big)\quad\text{and}\qquad |z\rangle = -i\,|0\rangle\,.$$

We see that $S^x|x\rangle{=}S^y|y\rangle{=}S^z|z\rangle{=}0$, and that these basis states are invariant against time reversal operation. The action of the spin operators in this basis can be written in a compact form

$$S^\alpha|\beta\rangle = i\sum_{\gamma=x,y,z}\varepsilon_{\alpha\beta\gamma}|\gamma\rangle\ .$$

[9] What exactly this field is depends on the situation.

Now, every hermitian operator O can be written as

$$O = \sum_{\alpha,\beta=1}^{3} A_{\alpha,\beta} \, |\alpha\rangle \, \langle\beta| \ ,$$

where $|1\rangle$, $|2\rangle$, $|3\rangle$ are basis states describing the $S = 1$ object (e.g., the ones defined above), and $A^*_{\alpha\beta} = A_{\beta\alpha}$, and which by the above construction is a quadratic form in the spin operators S^α. Such a self-adjoint $3{\times}3$ matrix is described by 9 real-valued parameters, from which we can introduce eight non-trivial, independent operators (the trace is held fixed, so that it is one parameter less than the number of entries in the matrix). These entries can be interpreted as possible on-site order parameters for systems, which are built up from $S = 1$ objects. More general, we can introduce a rank-k tensor operator $\mathbf{T}^k$, whose entries T^k_q satisfy the commutation relations

$$\big[S^z,\, T^k_q\big] = qT^k_q \quad \text{and} \qquad \big[S^\pm,\, T^k_q\big] = \sqrt{k(k{+}1) - q(q{\pm}1)}\, T^k_{q\pm1} \, .$$

From this, the 'highest' entry is a product of k operators S^+; for example, we obtain for $k = 1$ the components of $T^1_q = (S^+,\, S^z,\, S^-)$, which, because $S^\pm = S^x \pm iS^y$, are the three order parameters for local magnetizations in the three spatial directions (dipolar order). For $S = 1/2$, this exhausts all possibilities, since there we can apply the ladder operators $S^\pm$ maximally once, before the result is zero, i.e., $\mathbf{T}^2 = 0$ for $S = 1/2$ systems. However, for $S = 1$ this is not true, and for $k = 2$ we find

$$\begin{aligned}
T^2_2 &= S^+S^+ & T^2_{-2} &= S^-S^- \\
T^2_1 &= -\big(S^+S^z{+}S^zS^+\big) & T^2_{-1} &= \big(S^-S^z{+}S^zS^-\big) \\
T^2_0 &= \sqrt{\frac{2}{3}}\big(3(S^z)^2{-}S(S{+}1)\big).
\end{aligned}$$

The elements T^2_q are not automatically hermitian. Since the order parameter is an observable and hence hermitian, we can use the T^2_q to form suitable linear combinations, which then can be interpreted as local order parameters. We thus obtain the 5-component order parameter for spin-nematic or spin-quadrupolar order:

$$\mathbf{Q} = \begin{pmatrix} Q^{x^2-y^2} \\ Q^{3z^2-r^2} \\ Q^{xy} \\ Q^{yz} \\ Q^{zx} \end{pmatrix} = \begin{pmatrix} (S^x)^2 - (S^y)^2 \\ \frac{1}{\sqrt{3}}\big(2(S^z)^2{-}(S^x)^2{-}(S^y)^2\big) \\ S^xS^y + S^yS^x \\ S^yS^z + S^zS^y \\ S^zS^x + S^xS^z \end{pmatrix} .$$

The components of $\mathbf{Q}$ are called quadrupolar order parameters. Their expectation value can be finite also for states, which are invariant under time-reversal symmetry, for which the dipolar (magnetic) order parameters have zero expectation value. Hence, this construction lead us to a new type of order parameter, which is beyond the 'standard' view onto magnetic systems, in which only the dipolar (magnetic) order parameters are considered.

An alternative way to obtain the quadrupolar order parameters is the following: The quadratic form $A_{\alpha,\beta} = S^\alpha S^\beta$ is decomposed into a scalar $S(S{+}1)\delta^{\alpha\beta}/3$ (trace – corresponding to the length of the spin), a three-component vector $(S^{\alpha\beta}{-}S^{\beta\alpha})/2$ (antisymmetric representations: dipolar operators, pointing to a certain direction and thus breaking time-reversal symmetry – corresponding to the local magnetization), and a symmetric, traceless, tensor of rank two, $(S^{\alpha\beta}{+}S^{\beta\alpha})/2 - S(S{+}1)\delta^{\alpha\beta}/3$, corresponding to the five quadrupolar operators. How many local order parameters one can obtain in a quantum magnetic system then depends on the value of the spin S and, in general, the underlying symmetry, which typically for spin systems describing electronic systems is SU(2), but can be larger, SU($N{>}2$), as described in the introduction.
Note that a similar construction can be done for bond-order [82]: when summing the spins on neighboring bonds, then even for a $S = 1/2$ system we obtain $S_{Bond} > 1/2$, and a similar construction for higher-order order parameters can be done. The local order parameters for quadrupolar order then live on the bonds rather than on the sites, and hence this type of order is also called 'bond-nematic order'.
The number of possible order parameters is determined by the extent, to which one can apply the ladder operators $S^\pm$ without getting zero. For the SU(2) case, we see that for $S = 1$, similarly to the $S = 1/2$ case, we have $T_q^{k>2} = 0$, so the local magnetization and the quadrupolar order exhaust the possibilities to construct local order parameters related to the spin degrees of freedom. However, for $S > 1$, again further order parameters can be realized, named multipolar states of degree $k \leq 2S$, and the order parameters are rank-k tensor operators.
Since the so-constructed local order parameters rely on symmetry considerations, one can ask for possible relations between them. Indeed, one finds for the SU(2), $S = 1$ case

$$\langle \mathbf{S} \rangle^2 + \langle \mathbf{Q} \rangle^2 = \frac{4}{3}\,,$$

and that any state $|S,\ S^z\rangle$ is an eigenstate of $\mathbf{Q}^2$ for any spin S,

$$\mathbf{Q}^2\,|S,\ S^z\rangle = \frac{4}{3}S(S{+}1)\Big(S(S{+}1) - \frac{3}{4}\Big)\,|S, S^z\rangle$$

and hence

$$\left(\mathbf{Q}^2 + \mathbf{S}^2\right)|S, S^z\rangle = \frac{4}{3}S^2(S{+}1)^2\,|S, S^z\rangle\,.$$

2.1.2 How to identify long-range order

As mentioned before, the order parameter is finite only in the thermodynamic limit. The question arises, how to compute it in practical calculations, which often imply finite size systems. To do so, one can investigate the behavior of two-point correlation functions,

$$C(x, x') = \left\langle \psi \left| \mathcal{O}^\dagger(x)\mathcal{O}(x') \right| \psi \right\rangle\ .$$

Typically, one encounters the following behavior:

$$\lim_{|x-x'|\to\infty} C(x, x') \propto \begin{cases} \langle \mathcal{O}^\dagger(x)\rangle\langle \mathcal{O}(x')\rangle = \textit{const.} & \text{long-range ordered} \\ e^{-|x-x'|/l} & \text{disordered,} \end{cases} \tag{13}$$

with l the *correlation length*. In the presence of long-range order (LRO), the spatial average of the local order parameter will be finite: assuming locality,[10] the correlation function in Eq. (13) *factorizes*, and hence $C(x, x')$ approaches a constant, finite value when increasing the distance $|x - x'| \to \infty$. This corresponds to a divergent correlation length l, since the values of the order parameter at two points in space are correlated to each other for arbitrary separations between them. What is interesting is that, while the order parameter $O(x) = \langle \mathcal{O}(x) \rangle$ for a finite system is exactly zero, the two-point functions *can* show a finite value for *finite,* large enough separations! This opens the door to investigate LRO by working on finite systems by making sure that one treats systems large enough so that over a wide range of distances larger than a typical length scale determined by the details of the system, $|x-x'| > l_c$, the value of $C(x, x')$ is constant. This is, in particular, important for numerical approaches, where often one works with a finite lattice size.

Note that correlation functions can also decay to zero *algebraically*, $C(x, x') \propto |x-x'|^c$, with some exponent c. In this case, one speaks of *algebraic* or quasi long-range order, since the order parameter will have zero expectation value also in the thermodynamic limit. This happens, in particular, for low dimensional systems like spin-1 chains due to the *Hohenberg-Mermin-Wagner* theorem, according to which systems with short-range interactions in low dimensions cannot realize SSB of a continuous symmetry (essentially because fluctuations are too large). Therefore, in this case, the phases are not characterized by a finite order parameter, but by the *dominant* correlation function, i.e., the one, which decays *slowest*. An interesting scenario is realized in quantum magnets in so-called *Berezinskii-Kosterlitz-Thouless* (BKT) phase transitions: here, on one side of the transition, $C(x, x') \propto \exp(-|x-x'|/l)$, while on the other side of the transition $C(x, x') \propto |x-x'|^c$. On both sides of the transition, the order parameter is zero, but nevertheless the physics is different. Because one cannot analytically continue a power law to an exponential function, one encounters a real, thermodynamic transition between two states of matter, where $C(x, x')$ is nonanalytic at the critical point. This type of transition is not due to SSB, since the order parameter is zero on both sides. One example system for such a BKT transition is the classical XY-model on a square lattice, where as a function of temperature the binding or unbinding of *topological defects* causes this transition. Such an effect can be expected in XY-type models or models with U(1) symmetry. For quantum magnets, one often speaks of a BKT-type transition, if a gapless phase (with algebraically decaying correlation functions) is connected to a gapped phase (with exponentially decaying correlation functions), and at which the gap opens exponentially slowly: the transition is continuous, but in this case there is no thermodynamic potential (e.g. the free energy, which at zero temperature is the ground state energy), whose n-th derivative is nonanalytical, which is required by Ehrenfest's classification of a phase transition to be of n-th order. Therefore, one sometimes speaks of 'infinite-order transitions'.

[10] This is also referred to as the *cluster decomposition theorem*, according to which measurements of observables 'distant enough' from each other should be independent of each other.

2.1.3 What can be learned from correlation matrices

Let us have a closer look at the correlation function introduced in Eq. (13) (see, e.g., [84]). Let us switch from the continuous variable x to lattice positions i, j, so that we consider a correlation function $C_{i,j} = \langle \mathcal{O}_i^\dagger \mathcal{O}_j \rangle$. This is an hermitian matrix, and it is a valid question to ask, what we may learn from its eigenvectors and eigenvalues. Indeed, they give a valuable tool to investigate LRO in the following way: Let us consider the global order parameter $O = \sum_j O_j$. In the presence of translational invariance, we can write $O_j \equiv O/N = \bar{O}$. According to Eq. (13), we can write

$$\lim_{|i-j|\to\infty} \langle \mathcal{O}_i^\dagger \mathcal{O}_j \rangle = \overline{O}^2 \neq 0. \tag{14}$$

Let us consider a concrete example and choose $\mathcal{O}_j = S_j^+ = S^x + iS^y$, which addresses the question for finite in-plane magnetization. We see immediately, that (if S^z_{total} is a good quantum number) $\langle \psi | S_j^+ | \psi \rangle = 0$ for any finite system. However, $C_{ij} = \langle \psi | S_i^+ S_j^- | \psi \rangle$ can take a finite value even for small lattice sizes. Diagonalizing the hermitian matrix C_{ij} for such a finite system, we obtain real eigenvalues λ_ν and eigenvectors $\mathbf{v}_\nu$, and we can rewrite

$$C_{ij} = \sum_\nu \mathbf{v}_\nu \underbrace{\left\langle \psi \left| \left(\sum_i v^*_{\nu,i} S_i^+ \right) \left(\sum_i v_{\nu,j} S_j^- \right) \right| \psi \right\rangle}_{\lambda_\nu} \mathbf{v}_\nu^\dagger .$$

Introducing operators $\eta_\nu = \sum_i v_{\nu,i} S_i^-$, we obtain

$$\mathbf{v}_\nu^\dagger C_{ij} \mathbf{v} = \langle \psi | \eta_\nu^\dagger \eta_\nu | \psi \rangle = \lambda_\nu \geq 0 ,$$

the eigenvalues are therefore strictly positive. How does this relate to the order parameters? Let us apply the cluster decomposition theorem, then

$$\lim_{|i-j|\to\infty} C_{ij} = \lim_{|i-j|\to\infty} \sum_\nu v_{\nu,i} \underbrace{\left\langle \psi \left| \sum_k v^*_{\nu,k} S_k^+ \right| \psi \right\rangle}_{\sqrt{\lambda_\nu}^*} \underbrace{\left\langle \psi \left| \sum_l v_{\nu,l} S_l^- \right| \psi \right\rangle}_{\sqrt{\lambda_\nu}} v^\dagger_{\nu,j} = \lim_{|i-j|\to\infty} \langle S_i^+ \rangle \langle S_j^- \rangle .$$

However, the last equality can only be true if there is only *one* eigenvalue $\lambda_\nu = \lambda_{max}$, which is not vanishing. On the other hand, we see that $\langle S_j^+ \rangle = \sqrt{\lambda_{max}}$. For finite systems, these relations are not exact, but in the limit of infinite system size, there has to be asymptotically one dominant eigenvalue λ_L, so that for large systems of size L one can approximate

$$\lim_{|i-j|\to\infty} C_{ij} \approx \lambda_L v_{L,i} v^*_{L,j} .$$

Since the eigenvectors $\mathbf{v}_\nu$ are normalized, their coefficients scale $\sim 1/\sqrt{L}$. Hence, in order to have a finite value in the thermodynamic limit, the dominant eigenvalue has to scale $\lambda_L \sim L$, giving a condition on the largest eigenvalue of C_{ij}, which can be tested numerically.

Fig. 4 shows an example for the kagome lattice in the presence of DM interactions and of a non-magnetic impurity (see Fig. 2), for which this analysis was performed. For this system it

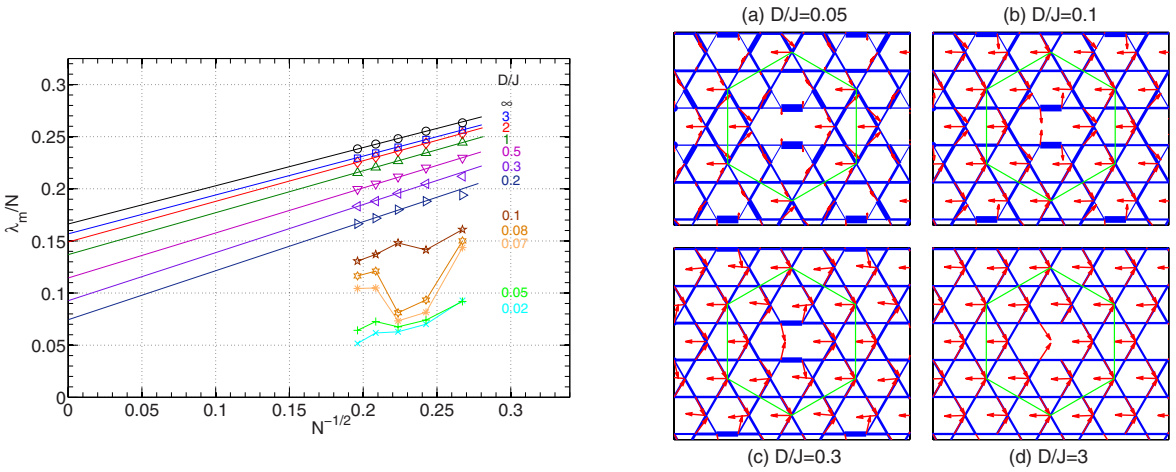

Fig. 4: *Correlation matrix analysis of the local magnetizations for the AFM Heisenberg model on a kagome lattice with DM interactions. Left: scaling of the largest eigenvalue of C_{ij} with system size. Right: Magnetization profile obtained from the eigenvector of C_{ij} belonging to the largest eigenvalue for different values of the DM interaction. The red arrows indicate the in-plane moments, given by the real and imaginary part of the entries of the eigenvector, respectively; the blue lines are the local bond strengths, which are computed separately (Figures taken from [74]).*

is very difficult to treat large system sizes due to the lack of symmetries. Nevertheless, using this analysis, it is possible to gain information on the possible LRO realized in the thermodynamic limit, which illustrates that this approach to computing LRO is suitable also for difficult situations.

2.2 Symmetry protected topological phases in quantum magnets: the AKLT state

As we have seen, states of matter are usually characterized by the Landau paradigm, in which a continuous phase transition and the associated phases are obtained by the SSB of one (or more) symmetries of the Hamiltonian and the emergence of a *local* order parameter [85]. This paradigm has been *the* framework for understanding phases of matter and phase transitions, until in the 1980s experiments discovered the integer [86] and later the fractional quantum Hall effect [87, 88]. These systems possess transitions between states with different conductivities, which apparently are not associated to any SSB. Subsequently, and also motivated by the discovery of high-temperature superconductivity [2], a new type of 'order' was proposed whose phenomenology is not due to the finiteness of some local order parameter, but in which the phases are characterized by *global* characteristics, like the degeneracy of the ground state or entanglement of the system. This type of order has been coined *topological order* [89,90,33] since the behavior is captured by topological field theories [91]. The main characteristics of topologically ordered phases are the presence of degenerate ground states, of gapless edge states, and the characterization in terms of *topological invariants* which are integer numbers capturing 'topological' properties of the system and which vary in the different phases. One characteristic of

topological phases is that they do not change under continuous deformations of the system (i.e., the topological invariant does not change unless one hits a critical point at which the system experiences singular behavior) and are, hence, protected against local perturbations like, e.g., noise. This makes these states very interesting for quantum computation in which one of the biggest challenges is to protect the entanglement between qubits from decoherence effects due to local noise induced by the environment. This approach is coined *topological quantum computation* and is described in the review article [33]. A lot of research is, therefore, devoted to uncovering such topological phases in quantum magnets.

At the present, obtaining a complete characterization of topological phases (including interacting systems) is an ongoing topic of research. However, one can use the following approach to distinguish between different gapped phases [92, 93]: a gapped quantum phase is characterized by ground states of Hamiltonians, which can be smoothly deformed into each other without closing the gap. An illustrative example is the $S = 1$ BLBQ chain (11), which at zero magnetic field displays a gapped phase for $-\pi/4 < \theta < \pi/4$; even though at $\theta = 0$ the Hamiltonian looks much simpler, the system in this parameter range is in the same phase since the gap closes only at the endpoints of this region. This property can be rephrased by saying that two ground states belong to the same phase if they are related by a *local* unitary transformation. Since local unitary transformations can only change local entanglement properties but not global ones, states in the same topological phase are characterized by the same 'long-range entanglement'. Based on these considerations, the following gapped phases can be identified:

1. Phases with 'short-range entanglement':

   (a) Topologically 'trivial' product states.

   (b) Symmetry protected topological phases (SPT). In these phases, local unitary transformations exist which preserve the symmetry of the state. Short-range entangled phases in which such a symmetry is broken are well described by Landau theory. Note that phases without local order parameter can still belong to different SPT phases if they are characterized by different symmetries, even though in Landau classification they would belong to the same 'disordered' phase.

2. 'True' topological order with 'long-range' entanglement, existing only in spatial dimensions $D \geq 2$ [92]. These phases are characterized by *anyonic* fractionalized excitations, which obey a generalized quantum statistics and are neither fermions nor bosons [33].

It is possible to characterize topological order by considering entanglement properties [94, 95], and tensor-network approaches have been introduced (see, e.g. [96] and the viewpoint [97]). While it is possible to investigate for 'true' topological order in 2D using the DMRG (for studies on the kagome lattice see, e.g. [35, 37, 36]), often SPT phases are investigated, which, according to the above said, are the only type of topological phases encountered in 1D. A prototypical example for an SPT state in quantum magnets is the so-called AKLT state [98] (named after the authors of the original publication, Affleck, Kennedy, Lieb, and Tasaki), which is depicted in

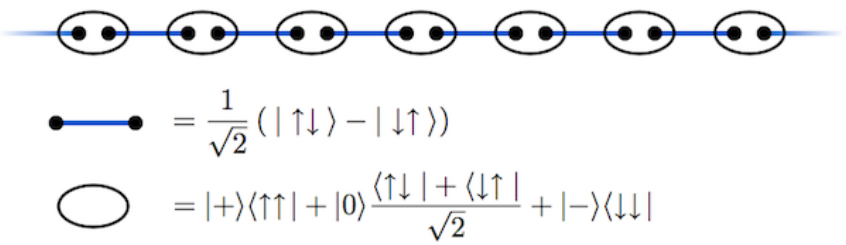

Fig. 5: *Sketch of the AKLT state (figure taken from Wikipedia).*

Fig. 5. It is the ground state of the BLBQ model (11) for $B = 0$ and $\tan(\theta) = 1/3$. It has several peculiar properties: the spin-1 degrees of freedom on the lattice sites are understood as being composed of two spin-$1/2$ degrees of freedom, which *between* neighboring lattice sites form singlets. At the edges, effective, free spin-$1/2$ degrees of freedom remain and form edge states – the spin-1 degrees of freedom 'fractionalize' to the 'smaller' spin-$1/2$ degrees of freedom. Since the presence of such edge states is typical for an SPT phase, one can use this to identify and characterize such phases, as seen further below.

SPT phases can numerically be detected by identifying an excitation gap, zero local order parameters, and degeneracy of the *entanglement spectrum* [99]. As discussed in [100, 95], in an SPT phase all states of the entanglement spectrum are non-trivially degenerate due to the symmetry in the system. Other indications for topological properties can be obtained from diagonalizing transfer matrices from which one can obtain directly the projective representations of the symmetry group [101], which can be used to further characterize SPT phases (see also [102] for a nice discussion of this aspect). According to [102–104], it is possible to distinguish between different SPT phases by applying the corresponding *active operators* on the edge states: if the correct active operator is coupled to the edge of the system, the ground state degeneracy is lifted. This can indeed be used to distinguish the different SPT phases in quantum magnetic systems obtained from the projective representations [105]. Numerically, it is easily seen that applying the 'wrong' active operator does not lift the ground state degeneracy, while applying the correct one leads to different energies of the ground states with a difference far greater than the numerical accuracy.

2.3 Dynamical properties: inelastic neutron scattering, electron spin resonance

One way to characterize a state is to weakly perturb it and to monitor its response. In this way, information beyond the LRO in the system can be obtained, and an insight about the elementary excitations in the system can be obtained. This is the realm of linear response theory. To study such a situation is interesting from many points of view. For example, the aforementioned spin liquids do not show any sign of LRO. Any investigation based on this will, therefore, not find any interesting properties and overlook that we are, indeed, facing a very rich quantum state of matter. However, the response of such states to weak perturbations is determined by the interactions in the system, hence these type of experiments are helpful for gaining insight into the true nature of the system.

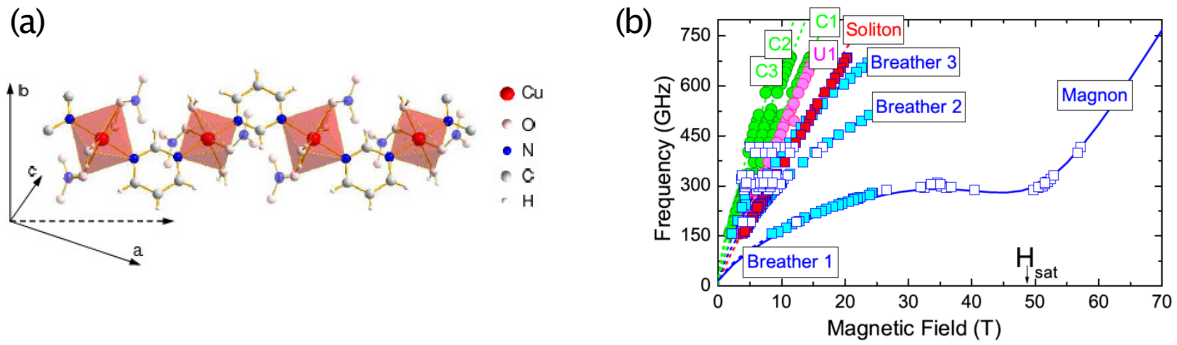

Fig. 6: *(a) Crystal structure of Cu-PM (copper pyrimidine dinitrate), a $S = 1/2$ spin chain material with DM interactions and alternating g-tensor. (b) Comparison of ESR spectra (symbols) and DMRG results (solid line) (Figures taken from Ref. [106]).*

From the experimental side, inelastic neutron scattering is one of the most important, direct probes for dynamical properties of quantum magnets. Since the neutron is charge neutral, one can perform the investigation such that only magnetic degrees of freedom are probed. The coupling of the magnetic moment of the neutron to the magnetic moments in the material allows one to measure the *dynamical structure factor*, which can be defined via

$$S^{\alpha,\alpha}(k,\omega) = \frac{1}{N}\sum_{j=1}^{N} e^{-ik(j-N/2)} \int_{-\infty}^{\infty} dt\, e^{i\omega t} \left\langle S_j^{\alpha}(t) S_{N/2}^{\alpha}(0)\right\rangle \,.$$

Here, the system contains N spins, $\alpha = x, y, z$, and we assume translational invariance. These experiments can be performed also at high magnetic fields, so that the excitation spectrum of quantum magnets for different phases can be investigated.
Another interesting type of experiments are electron spin resonance experiments (ESR), which give access to the imaginary part of the dynamical structure factor in the long wavelength limit $k \to 0$. An example for the ESR spectrum of a spin-chain material with DM interactions is shown in Fig. 6. As can be seen, different types of excitations (e.g. solitons, breathers) can be identified by comparing to an effective field theory (in this case a sine-Gordon theory).

3 Nonequilibrium dynamics

So far we considered static properties of quantum magnets, which uncover a multitude of interesting phenomena. It is only natural to ask what happens if one now excites such a system, e.g., with a laser pulse, so that its state evolves in time. The typical questions one can ask is how the order parameters decay in time, and what nature the equilibrium state will be. Also, one can ask the reverse question: is it possible to induce order (e.g. magnetization) by exciting the system? These are recent topics of study and in this final section of this book chapter we give a short glimpse on some of the ongoing developments.
In the mid 1990s, investigations were performed on thin magnetic films, which were excited with an ultrashort laser pulse of duration 60 fs (see, e.g., [107] for a review). The magnetization was measured using the magneto-optical Kerr effect (MOKE), and, since the signal was

thought to be proportional to the magnetization, they were able to measure the time evolution of the magnetization after such a short excitation. Interestingly, they found that the magnetization decreased very quickly (on a time scale < 1 ps), but then recovered again. Later, it was argued that the MOKE signal is not necessarily proportional to the magnetization in such a nonequilibrium situation, but these results were reproduced using other techniques. This behavior raises many questions. One main point is to understand where the angular momentum, which is underlying to the magnetization, dissipates to, and how the light-matter interaction triggers the dynamics. Typically, a *three temperature model* is introduced: the energy absorbed from the laser flows to (i) the electrons, (ii) the lattice vibrations, i.e., creation of phonons, and (iii) the spin degrees of freedom of the system, by creating magnons. Often, in the theoretical studies, time-dependent density functional theory is applied. However, despite now about 30 years of research, no consensus has been reached on the mechanism of the demagnetization dynamics [107]. One possibility would be to address many-body spin systems like the ones described in this book chapter; however, since the electron and phonon degrees of freedom also seem to play an important role in the magnetization dynamics, one would need to extend the models correspondingly. Treating such complicated many-body models is a challenge for ongoing and future research. However, it would be interesting to see if other types of order, e.g., spin-quadrupolar order in spin-1 systems, could show similar time dependence, or if the demagnetization dynamics only affects magnetic order.

In materials with two different types of magnetic ions (e.g. Heusler compounds), optical excitation can lead to an effective transfer of spins from one atomic species to the other [108]. This effect was coined 'optically induced spin transfer' (OISTR) [109, 110] and builds on the observation that, even in the presence of SOC (which, as discussed in Sec. 1.3, does not preserve S^z as good quantum number) on very fast time scales $\lesssim 11$ fs no spin flips happen. Instead, the spin is simply transferred to the neighboring ion, leading to a change in the magnetization pattern, and the charge distribution on the lattice. At later times, SOC may come into play and may cause demagnetization dynamics, but at least on the very short time scale the OISTR mechanism leads to a metastable or transient state, which is different from the initial state. Such a situation can also be studied in Hubbard-type models with an underlying magnetic microstructure, where OISTR leads to the weakening of the original spin structure, but induces charge density wave type structures, which prevail until further effects like SOC or phonons come into play [111].

Other interesting effects when going out-of-equilibrium are the possibility to realize (transient) long-range order. A famous example is the description of transient superconductivity in pump-probe experiments [112]. Also, the formation of magnetic LRO has been reported [113]. For example, in the manganite material $GdSrMnO_3$, a photo-induced transition to a ferromagnetic metallic phase within 200 fs has been observed [114] and can be described by first-principles approaches [115]. It will be interesting to further study the possibility to realize (transient) LRO in quantum magnets, in particular regarding the realization of the unconventional states discussed in this contribution. Maybe some of the LRO that is possible in interacting spin systems, but hard to realize in ground states, can be found in such nonequilibrium setups in future investigations.

4 Conclusions and outlook

Quantum magnetism is a vast field of research, and in this contribution only some aspects could be discussed. What remains appealing is the possibility to identify by mathematical considerations further order parameters, which can help to identify novel types of LRO, for which the spin-nematic order is one example. The newer developments for nonequilibrium systems are a promising way to go, since there many basic questions on the nature of transient order are still under investigation. It will be interesting to see whether in such situations novel behavior can be identified, and if it will be long-lived, so that it does not vanish in less then a blink of the eye, but can be enjoyed on a useful time scale.

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