Quantum magnetism in low dimensional spin systems

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Low-dimensional quantum magnets provide a unique possibility to study ground and excited states of a large variety of quantum models with very high accuracy [1]. Despite their conceptual simplicity, these models provide very rich magnetic phase diagrams [2, 3] arising from a complex interplay of exchange interactions and magnetic frustration which often result in strong quantum fluctuations. In particular, dimensionality and lattice topology are decisive for the observed plethora of quantum phenomena like the realization of a Tomonaga-Luttinger liquid in $YbAlO_3$ [4-6] the possible quantum spin liquid state in the Cu-mineral Barlowite [7], the formation of skyrmions in Cu_2SeO_3 [8-10] or the field induced magnetization plateau in Boleite.

$YbAlO_3$ – a realization of a Tomonaga-Luttinger liquid

In condensed matter physics, among the large number of well-studied theoretical models there are some that have very rare experimental realization. This is valid even for the simplest models like the one-dimensional (1D) antiferromagnetic (AFM) quantum spin ½ chains. This model can be well described by the exotic Tomonaga-Luttinger liquid (TLL) theory and exhibits exceptional properties, such as strong quantum fluctuations and the presence of so-called "spinons". In conventional 3D magnets, in which the AFM ground state consists of a periodic arrangement of magnetic moments, an elementary excitation can be represented as a wave of small deflections of these moments from their initial position, or as a quasiparticle known as magnon which carries spin momentum S = 1. On the other hand, in the 1D case the situation is different: a magnon is "fractionalized" into two fermionic quasiparticles known as "spinons".

The process of creation of a spinons pair is schematically illustrated in Fig. 1. These spinons can propagate freely (deconfined) along the AFM chain at temperature above the ordering temperature T_N (Fig. 1 b,c) while the presence of an effective internal field due to the intrachain interactions at $T < T_N$ confines the spinons (Fig. 1 d) because their propagation will cost energy.

We have recently demonstrated that, in the rare-earth quantum magnet $YbAlO_3$ (with an AFM ordering temperature T_N = 0.88 K, see Fig. 2), the TLL is experimentally realized [1]. This was done by a combination of thermodynamic (at MPI-CPfS in Dresden) and spectroscopic (at Oak Ridge National Laboratory) measurements. The low-temperature magnetization measurements revealed a very strong uniaxial anisotropy of the Yb moments, which is induced by a combination of spin-orbit coupling and

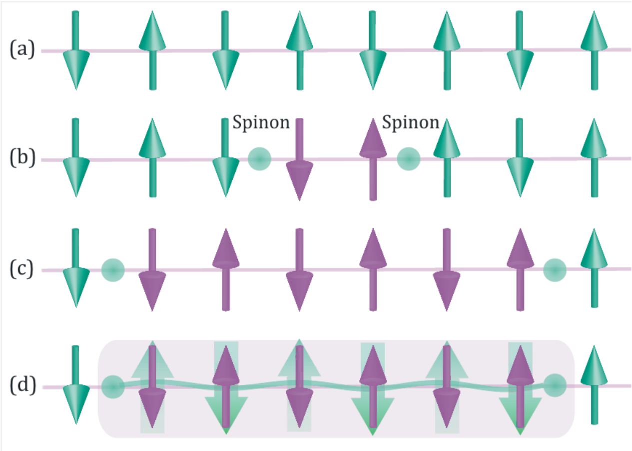

Fig. 1: Sketch of the spinon creation in an AFM spin chain: (a) ground state of the AFM chains is an ordered pattern of alternating spin moments. (b,c) elementary excitation (a flip of two neighbor spins) creates two spinons, which can propagate freely along the chain (iv) Below the ordering temperature the presence of the effective magnetic field from the neighbors chains (shown by thick arrows) increases the energy cost of the spinons © MPI CPfS

crystalline electrical field effect [2]. On the other hand, results of inelastic neutron scattering above the ordering temperature show a gapless spinon continuum, dispersive along the direction of the Yb chains, indicating that the collective magnetic behavior is dominated by the highly isotropic Yb-Yb exchange interaction and forms a TLL state. This is indicated in Fig. 2 which shows the magnetic field - temperature phase diagram of $YbAlO_3$ with field along the crystallographic a-axis. Note that the combination of highly anisotropic magnetic moment of Yb and isotropic Yb-Yb exchange interaction is among the unique properties of $YbAlO_3$ giving a rise to its unusual behavior and rich excitation spectra.

With decreasing temperature, a dipole-dipole intrachain interaction confines the spinons and induces

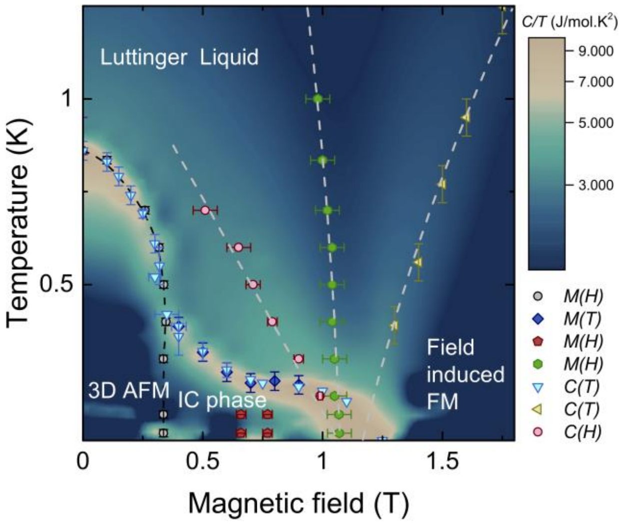

Fig. 2: Experimental magnetic field - temperature phase diagram of $YbAlO_3$, with magnetic field applied along the crystallographic a-axis. With increasing field the AFM order changes into a spin-density-wave (SDW) and transverse antiferromagnetic (TAF) phases. At B = 1.15 T a quantum critical point (QCP) is found.

a simple long-range AFM ordering of the Yb moments. The application of a magnetic field, at first, creates an exotic longitudinal spin-density-wave (SDW) phase, and with further increasing, destroys the long-range order at the *quantum critical point (QCP)*. Most remarkably, thermodynamic properties of $YbAlO_3$ near the *QCP* follow a universal behavior and the analysis of the scaling behavior and critical exponents indicates that the *QCP* is a free fermion fixed point, consistently with expectation for the 1D TLL. More details can be found in Refs. [1-3].

Boleite – an Archimedian Solid with strong magnetic frustration

The beauty of brightly colored and highly symmetric mineral specimens has fascinated humankind for millennia. In addition to these characteristics, the deep-blue cubes of the mineral Boleite (see Fig. 3) exhibit an Archimedean solid crystal lattice of magnetic spin-1/2 Cu-atoms arranged on well separated truncated cubes (see inset Fig. 4), connecting this way mineralogy, crystallography, chemistry, mathematics and quantum physics. Our study, combining theoretical and experimental efforts, provides a microscopic understanding of the strongly frustrated quantum-magnetism of Boleite, which turns out to exhibit a unique example of a molecular-like magnet in a three-dimensional solid. We demonstrate how the combination of geometrical frustration, isotropic and anisotropic magnetic exchanges, and the concept of

Fig. 3: Natural light blue and transparent Boleite crystals from Antofagasta, Chile (image width: 2mm)

randomness to describe defects, leads to a consistent microscopic description of the unusual magnetic properties of Boleite.

In our combined experimental and theoretical study, we apply density functional (DFT) electronic structure calculations and subsequent exact diagonalization (Finite temperature Lanzos Method, FTML) for the derived model Hamiltonian to understand the measurements in high magnetic fields up to 60 Tesla. Since Boleite is a naturally grown material, great care is taken with respect to the selection and characterization of the samples. As main result, we find that the magnetism of Boleite with its peculiar 1/3-plateau in high magnetic fields can be treated in a very good approximation as frustrated molecular quantum magnets assembled in a solid. The influence of intrinsic imperfections in the naturally grown Boleite crystals, modifying the ideally expected behavior, can be well understood from our simulations.

To estimate the relevant magnetic exchange couplings, we start with a density functional electronic structure calculation. We use the experimental crystal structure, but optimize the hydrogen positions with respect to the total energy, since in the available experimental data the H positions are often ill defined or even unknown. For the calculation of the exchange couplings, however, accurate H positions are crucial.

Being well aware of sample issues in minerals, in particular problems of non-stoichiometry, epitaxy and isomorphism in the Boleite mineral group, we attempted to find crystals of the highest quality from different localities. Indeed, only samples from one locality (Antofagasta, Chile) showed the characteristic plateau in the field dependent magnetization expected from our model (see Fig. 4).

Combining a tight-binding model and total energy LDA+U calculations, we can evaluate three relevant exchange couplings: A ferromagnetic J_D=-75K for the dimers on the edges of the truncated cube, an antiferromagnetic J_T=240K for the trimers on the corners and a small next nearest neighbor coupling J_L=5K between them. To complete our microscopic model, we estimated the antisymmetric Dzyaloshinskii-Moriya (DM) anisotropy, which is the leading correction to the magnetic exchange due to the spin-orbit coupling. We find DM vectors whose magnitude amounts to about 0.06 of the respective isotropic exchange. Hence, for the magnetization process, the moderate DM anisotropy will play a minor role.

In order to rationalize the magnetic observables, we model the truncated cubes of Boleite by means of a spin model of independent 24-spin clusters (see inset Fig. 4). The interactions are of Heisenberg type. On the edge of feasibility, complete diagonalization studies of the Hamiltonian at arbitrary magnetic fields are accessible by the finite-temperature Lanczos method (FTLM). However, to obtain susceptibility and magnetization curves that describe the experimental data well, we have to introduce in our simulations the rather new concept of “exchange randomness” which describes the intrinsic imperfections of the crystals. Such distribution of the exchange parameters around the values derived for the perfect crystal structure leads to a significantly smoother dependence of susceptibility and magnetization from temperature and magnetic field, respectively.

Experimentally, Boleite features a characteristic 1/3-plateau (see Fig. 4) in the field-dependent magnetization M(H) as a signature for its highly frustrated magnetism. This is in strikingly good agreement with our theoretical modeling. Owing to the large distance and the respective weak coupling between the truncated cubes, the magnetism of Boleite can be treated in a very good approximation as very weakly interacting molecular magnets - with 24 Cu-spin 1/2-sites arranged on vertices of the Archimedean truncated cube - assembled in a three dimensional solid. Such regular bodies are of particular interest to study models for low-dimensional quantum magnetism, since these models are, like in our case, often solvable numerically exactly.

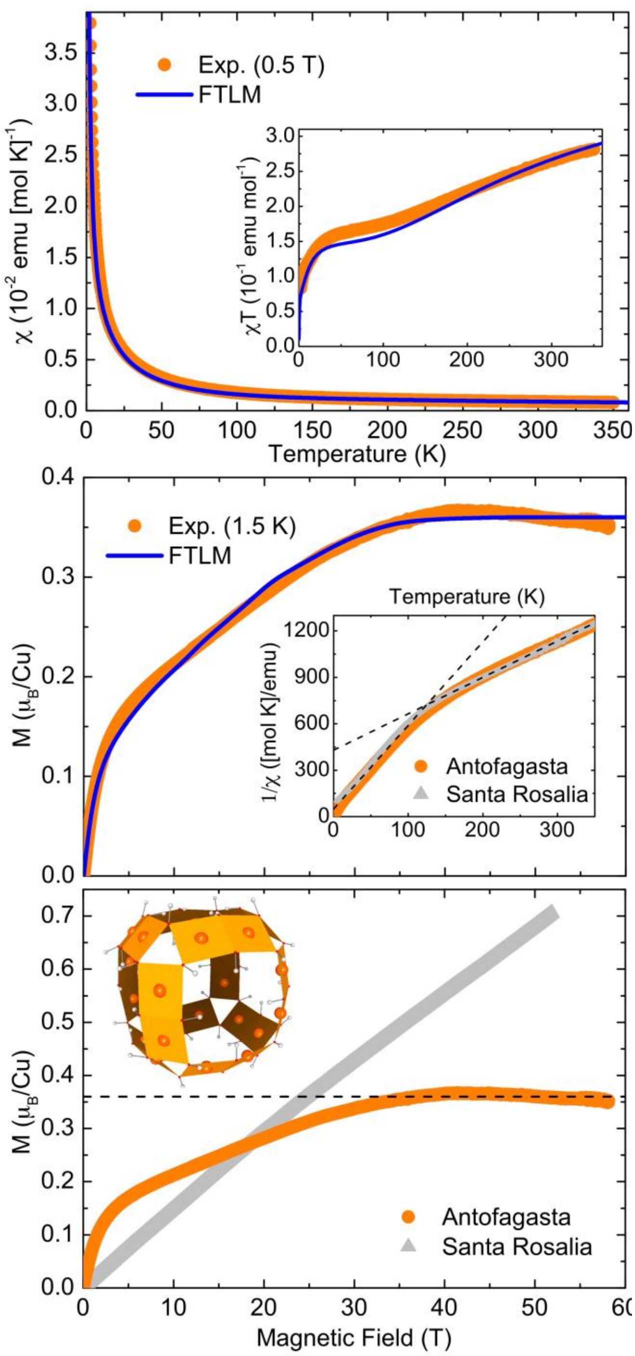

Fig. 4: Upper panel: Measured and calculated (FTLM) magnetic susceptibility using the DFT derived J’s and an additional exchange randomness. Middle panel: Measured and calculated high field magnetization for different samples. Lower panel: Only the transparent specimen from Antofagasta (Chile) show the expected characteristic 1/3 plateau at high fields. Inset: Scheme of the 24 Cu sites on the truncated cube in the crystal structure of Boleite.

External Cooperation Partners

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