K124176 OTKA grant
Magnetism, topology, and entanglement in quantum insulators

Final report

Our principal aim was the study of the magnetic and electric properties of correlated materials when quantum effects are significant. We have focused on Mott insulators when the degrees of freedom are localized. Examples include magnetic materials with spin degrees of freedom and ultracold atoms in optical lattices. The low-energy dynamics in such a system originates from the Heisenberg exchange, which, in the simplest case, swaps the local spin states of two neighbouring atoms. However, it can include other terms that arise from the particularities of the material. For example, the relativistic spin-orbit coupling introduces anisotropies like the antisymmetric Dzyaloshinskii–Moriya interaction or the symmetric Kitaev-like interaction, leading to nontrivial topological properties. The spin-orbit coupling is also responsible for the interaction between the magnetic moments and the electric polarisation in materials where the inversion symmetry is absent — this is the so-called magnetoelectric coupling. It leads to new phenomena in multiferroic materials with coexisting magnetic and electric orders.

Frustration, i.e. the inability of the spin to optimize all interactions simultaneously, is another source of fascinating behaviour. Frustrated spin systems may show spin-liquid behaviour with unconventional ground states and excitations. Spin liquids often appear in extreme quantum cases, as in models for ultracold atoms on optical lattices. But spin liquids may even appear for classical spin systems on frustrated lattices. We performed analytical and numerical calculations to address the phenomena described above.

In collaboration with experimentalists, we have shown that the magnetoelectric coupling in multiferroics with antiferromagnetic order parameters allows the manipulation of the antiferromagnetic domains by cooling the samples in external electric and magnetic fields. Furthermore, we have demonstrated that one can identify the different domains by measuring the absorption of far-infrared light. In another experimental-theoretical project, we discovered the negative thermal expansion in the spinel compound $CdCr_2O_4$. We describe these results in Sec. 1 below.

In Sec.2, we report on our results on the quantum systems with multiple (N) degrees of freedom per site described by the SU(N) Heisenberg model, relevant for ultracold atoms on optical lattices. We identified, among others, a chiral spin liquid ground state on the triangular lattice.

The effect of exchange anisotropies on the topological properties of spin excitations is discussed in Sec. 3. Notably, we have found that the magnons in the Kitaev model in the magnetic field are topological, with finite Chern numbers. We have also discussed the stability of a particular $Z2$ topological invariant in spin systems.

Sec. 4 deals with results concerning a type of spin liquid in classical spin models, the so-called “spiral spin liquids”, where the ground state ordering momenta span a sub-extensive manifold. We have provided a systematic recipe to construct spiral spin liquids on Bravais lattices.

In Sec. 5, we have collected some results using the continuous-time quantum Monte Carlo method. These deal with a different class of disordered systems. E.g. we have studied how quantum fluctuations melt the Coulomb glass of fermions. Furthermore, we have explored the dynamics of the quantum Sherrington-Kirkpatrick spin-glass and compared it to experimental results.

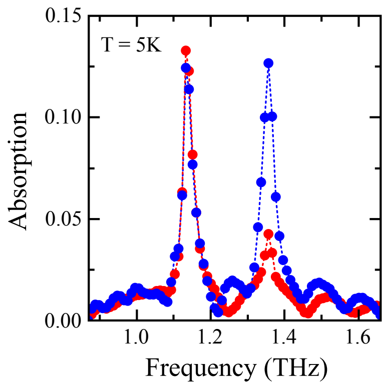
It is only a selection of our results since we published 32 articles in refereed journals within this project. These include 4 Physical Review Letters, 11 Physical Review B, 2 Physical Review A, 4 Physical Review Research, and 4 Quantum. The total impact factor is over 125, not counting the articles in the Physical Review Research, for which the impact factor is not yet available.

Two PhD students participated in the project: Péter Balla, who successfully defended his PhD thesis in February 2023, and Dániel Vörös, who is now in his fourth year of studies in the physics doctoral school at the Budapest University of Technology and Economics. In addition, three undergraduate students joined: Kondákor Márk, Péter Kránitz, and László Rudner. M. Kondákor and P. Kránitz carried out undergraduate research and presented their work at TDK conferences and as a poster at the Highly Frustrated Magnetism conference held in June 2022.

1. Results published in collaboration with experimentalists

Identification and manipulation of Antiferromagnetic Domains multiferroic materials
Phys. Rev. Lett. **121**, 057601 (2018) [arXiv:1711.08124];
Phys. Rev. Lett. **127**, 157201 (2021) [arXiv:2101.10045].

During the last couple of decades, the great potential of multiferroic materials in realizing magnetoelectric memory devices has led to the revival of the magnetoelectric effect and the search for multiferroic compounds. In multiferroics-based memory devices, the writing and reading of magnetic bits by electric field may be realized via the magnetoelectric coupling between the ferromagnetic and ferroelectric orders. However, ferro-ordered phases are extremely sensitive to external fields. As an alternative approach, information could be stored in antiferromagnetic domains, a concept proposed for metallic compounds in the framework of antiferromagnetic spintronics. We searched whether similar phenomena happen in insulators with coupled antiferromagnetic and antiferroelectric orders. We experimentally demonstrated that the magnetoelectric effect in $LiCoPO_4$ can be exploited not only for the control but also for the identification of antiferromagnetic domains via the strong directional dichroism detected in the THz frequency range -- the absorption coefficients in $LiCoPO_4$ were different for light propagating along and opposite to a given direction in the crystal. The origin of this phenomenon is the chirality of the light, as the electric and magnetic field components of the light and its propagation vector form a right-handed system. We identified the microscopic mechanism responsible for the magnetoelectric effect, which may also arise in other antiferromagnetic materials. The same principle may apply to the conventional optical imaging of antiferromagnetic domains in multiferroic materials with micrometer resolution. Furthermore, we demonstrated the in-situ electric control of the population of antiferromagnetic domains in another multiferroic material, $Ba_2CoGe_2O_7$. By cooling the sample in external magnetic and electric fields, we were able to control the population of the four symmetry-allowed domains in this material. The domains were identified by analyzing the NDD response.



Absorption of THz light in $LiCoPO_4$ at low temperatures. Cooling the crystal in perpendicular electric and magnetic fields of different signs selected one or the other antiferromagnetic domain. The difference in the absorption peak in different domains (red and blue curves) at 1.36 THz can be explained by the magnetoelectric coupling in the material.

Negative thermal expansion in the plateau state of a magnetically-frustrated spinel
Phys. Rev. Lett. **123**, 027205 (2019) [arXiv:1812.05890].

Materials usually expand upon heating: this is the usual thermal expansion. However, there are cases when a material displays "negative thermal expansion" (NTE) when it expands as it is cooled. In an experimental and theoretical collaboration, we reported on the negative thermal expansion in the high-field, half-magnetization plateau phase of the frustrated magnetic insulator $CdCr_2O_4$ at low temperatures (below 4K) and high magnetic field (at 30T). High-precision dilatometry measurements revealed a sizeable negative thermal expansion associated with the collinear half-magnetization plateau in magnetic fields above 27 T. This particular property of the $CdCr_2O_4$ is explained by the highly frustrated lattice the magnetic chromium ions form and the strong coupling between the lattice and the magnetic ions called magnetostriction, which helps the spin to satisfy their magnetic energy at the price of deforming the material. We provided a simple microscopic theory for spin-lattice coupling and identified the origin of the negative thermal expansion in the nearly-localized band of spin excitations in the half-magnetization plateau phase.

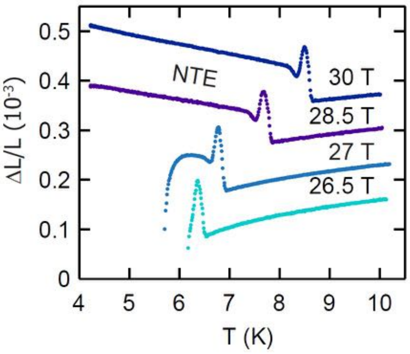


Figure: Thermal-expansion ($\Delta L/L$) measured from 4.2 to 10.4 K in magnetic fields up to 30 T, showing transitions from a paramagnetic state to a half-magnetization plateau state. The negative slope for the 28.5 T and 30 T curves shows the presence of NTE in the plateau state above 27 T.

2. Quantum insulators with multiple flavours

The search for unconventional states of matter like quantum spin liquid phases is an active field of research due to their fascinating physical properties and possible applications in quantum devices. Recent progress in experiments with ultracold atoms in optical lattices opens the exciting new possibility to simulate quantum models that potentially host such exotic quantum states. In particular, fermionic alkaline rare-earth atoms provide a nuclear spin degree of freedom which allows realizing SU(N) symmetric Hubbard models at strong coupling ($N=2I+1$, where I is the nuclear spin). The physical properties of these models are largely unexplored, which creates a strong motivation for theoretical investigations to name potential realistic hosts of quantum spin liquid phases.

The bilinear–biquadratic model on the complete and bipartite crown graphs
J. Phys. A **51**, 1751 (2018) [arXiv:1709.06602];
Phys. Rev. B **103**, 214448 (2021) [arXiv:2001.08310].

We studied the spin-1 bilinear-biquadratic model on the complete graph of N sites, i.e., when each spin is interacting with every other spin with the same strength. Because of its complete permutation invariance, this Hamiltonian can be rewritten as the linear combination of the quadratic Casimir operators of SU(3) and SU(2). Using group representation theory, we explicitly diagonalized the Hamiltonian and mapped out the ground-state phase diagram of the model. Furthermore, we presented the complete energy spectrum, including degeneracies, analytically for any number of sites. Next, we studied a bipartite collective spin-1 model with exchange interactions between the spins. The bipartite nature of the model manifests itself by spins being divided into two equal-sized subsystems, a setup inspired by recent experiments with ultracold atoms. Using the SU(3) symmetry of the exchange interaction and the permutation symmetry within the subsystems, we employed representation theoretic methods to diagonalize the Hamiltonian of the system in the entire parameter space of the two coupling strengths and construct the ground-state phase diagram. We got a rich phase diagram, containing both gapped and gapless phases.

Dimensional crossover in the SU(4) Heisenberg model in the six-dimensional antisymmetric self-conjugate representation: Quantum Monte Carlo versus linear flavour-wave theory
Phys. Rev. B **100**, 085103/1-8 (2019) [arXiv:1906.06938].

Using linear flavour-wave theory and auxiliary field quantum Monte Carlo, we investigated the properties of the SU(4) Heisenberg model on the anisotropic square lattice in the fully antisymmetric six-dimensional irreducible representation, a model describing interacting fermions with four flavours at half-filling. Quantum Monte Carlo calculations on very large systems convincingly demonstrated that results are consistent with a small but finite antiferromagnetic moment at the isotropic point. The presence of a long-range antiferromagnetic order has been further confirmed by showing that a phase transition takes place into a valence bond solid phase not too far from the isotropic point when reducing the coupling constant along one direction on the way to decoupled chains, possibly providing a new example for a deconfined quantum criticality scenario.

Time-reversal symmetry breaking Abelian chiral spin liquid in Mott phases of three-component fermions on the triangular lattice
Phys. Rev. Research **2**, 023098 (2020) [arXiv:1802.03179]

We provided evidence for spontaneous chiral symmetry breaking in the Mott phase of the SU(3) Hubbard model on the triangular lattice. The SU(3) Hubbard model with only nearest-neighbour hopping and on-site repulsion is very realistic - it contains the main parameters expected to pop up with cold atoms and only those. The chiral phase is stabilized in the Mott phase with one particle per site in the presence of a uniform π-flux per plaquette, and in the Mott phase with two particles per site without any flux. Our approach relied on the study of the effective spin models derived in the strong-coupling limit using exact diagonalization and variational Monte Carlo simulation, as well as exact diagonalization of the SU(3) Hubbard model on small clusters. Besides magnetically long-range ordered phases, we found a nematic phase breaking the lattice rotation symmetry, and a spontaneous time-reversal and parity symmetry breaking Abelian chiral spin liquid.

Dynamical structure factor in SU(3) and SU(4) spin-liquids
Phys. Rev. B **104**, 184426 (2021) [arXiv:2107.09588]

We implemented a variational Monte Carlo method to calculate excitations in SU(N) symmetric Heisenberg models. The variational method relies on a truncated Hilbert space spanned by the Gutzwiller projected particle-hole excitations of the Fermi sea. We benchmarked the method on the SU(3) Heisenberg chain: We computed the dynamical spin structure factor and compared it to exact diagonalization results for 18 sites and the two-soliton continuum of the Bethe ansatz for 72 sites, and we got an excellent agreement in both cases. Detailed analysis of the finite-size effects shows that the method captures the Wess-Zumino-Witten SU(3) level 1 critical behaviour and reproduces the correct exponent. We also calculate the single-mode approximation for the SU(N) Heisenberg model and determine the velocity of excitations. For SU(3) Haldane-Shastry model, we found that the variational method gives the exact wave function for the lowest-lying excitation in the dynamical structure factor. Following the one-dimensional study, we implemented the method to the 2-dimensional SU(4) Heisenberg model on the honeycomb lattice with algebraic decay of spin correlations. The manuscript is in preparation.

3. Topological excitations in magnetic systems

Topological Magnons in Kitaev Magnets at High Fields
Phys. Rev. B **98**, 060404(R) (2018) [arXiv:1802.04283]

Recently, the effects of spin-orbit coupling (SOC) in correlated materials have become one of the most actively studied subjects in condensed matter physics. Among candidate materials, iridium oxides and $RuCl_3$ are believed to realize the Kitaev spin liquids. This motivated us to investigate the Kitaev-Heisenberg model that describes the magnetism in these spin-orbit coupled honeycomb lattice Mott insulators. We considered the model in strong magnetic fields perpendicular to the plane of the lattice that brings the system into the polarized paramagnetic phase. We found that the spin-wave bands carry nontrivial Chern numbers over large regions of the phase diagram, implying the presence of chiral magnon edge states. In contrast to other topological magnon systems, the topological properties here stem from the presence of anisotropic terms in the Hamiltonian that do not conserve the number of magnons. Using time-dependent density matrix renormalization group methods and interacting spin-wave theory, we demonstrated the presence of the chiral edge mode and its evolution with the field. Since the effects of interactions are suppressed by the exchange scale divided by the applied field strength, the validity of the single-particle picture is tunable, making paramagnetic phases particularly suitable for the exploration of this physics.

Multipolar edge states in the breathing kagome model
Phys. Rev. B **99**, 014408/1-13, 2019 [arXiv:1801.07950]

We propose that unconventional excitations of magnetic insulators can not only realize the analogues of topological electron bands but may accommodate other novel topological states. To illustrate this, we used the example of the kagome antiferromagnet, extended with breathing anisotropy and Dzyaloshinskii–Moriya interaction. Starting from entangled trimers as the magnetic units of the theory, the excitations are characterized by a spin-1/2 doublet and a spin-3/2 quartet. In the chiral magnetic state, a small magnetic field removes the degeneracy of the multiplets, and the excitations become topologically nontrivial with finite Chern numbers. Increasing the magnetic field, the spin-3/2 quartet excitations undergo a band-touching topological transition when a spin-3/2 Dirac cone is formed. By the bulk edge correspondence, chiral edge modes appear for open boundary conditions in the gap of the topological multiplets. These edge modes carry the multipolar characters of the high-spin excitations, giving room for novel quadrupolar edge states.

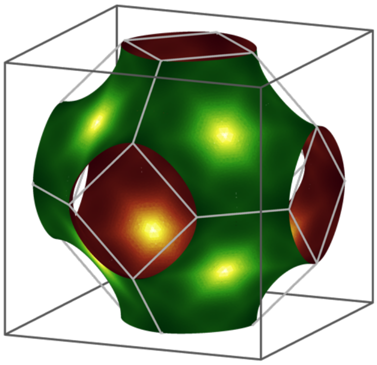
The fragility of Z_2 topological invariant characterizing triplet excitations in a bilayer kagome magnet
Phys. Rev. B **104**, 104412/1-22 (2021) [arXiv:2012.11765]

The discovery by Kane and Mele of a model of spin-1/2 electrons characterized by a Z_2 topological invariant had a lasting effect on the study of electronic band structures. Given this, it is natural to ask whether the band-like excitations of magnetic insulators may realize similar phenomena. Recently, models supporting Z_2 topological invariants have been proposed for both magnons and triplet excitations. In both cases, magnetic excitations form time-reversal (TR) partners, which mimic the Kramers pairs of electrons in the Kane-Mele model but do not enjoy the same type of symmetry protection. We revisited this problem in the context of the triplet excitations of a spin model on the bilayer kagome lattice. The triplet excitations provide a faithful analogue of the Kane-Mele model as long as the Hamiltonian preserves the TR × U(1) symmetry. We found that exchange anisotropies, allowed by the point group and typical in realistic models, break the required TR × U(1) symmetry and instantly destroy the Z_2 band topology. This suggests that the Z_2 topology of band-like excitations in magnets may be intrinsically fragile, in contrast to their electronic counterparts.

4. Frustrated classical spins

Degenerate manifolds in the classical Heisenberg model
Phys. Rev. B **100**, 140402(R), 2019 [arXiv:1905.12504]
Phys. Rev. Research **2**, 043278/1-25 (2020) [arXiv:1905.12504]

Even though the Heisenberg model for spins on face-centred-cubic lattices has been around for almost a century, a systematic study of the classical phase diagram for exchanges going beyond the second nearest neighbour was not done yet. Inspired to fill this gap, we studied the ground-state phase diagram of the model in detail, including both ferro- and antiferromagnetic exchanges. We found many interesting results on the way. Most remarkably, we identified points in the phase diagram where the ordering momenta form lines and surfaces in reciprocal space, leading to ground state manifolds with subextensive degeneracy. In reciprocal space, these degeneracies appear as manifolds of wave vectors, their dimensionality increasing with the degree of frustration and the robustness of the disordered spin-liquid state. We also presented a recipe to explicitly construct Heisenberg models on Bravais lattices (including the face-centred-cubic) with codimension-one manifolds, i.e., lines in two dimensions and surfaces with different Euler characteristics in three dimensions. Such ground states are known in the literature for the honeycomb and diamond lattices and are referred to as “spiral spin liquids”. We discussed the role of thermal and quantum fluctuations in stabilizing ordered states.

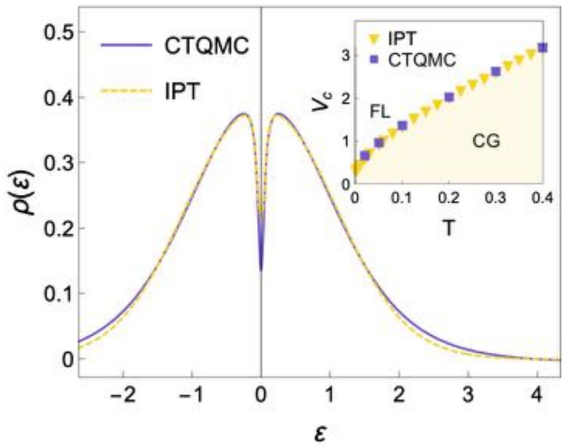


A ground state manifold of the 3-dimensional face-centred-cubic lattice in the reciprocal space. The surface is coloured according to the free energy, brighter colours correspond to states with smaller values of free energy. Thermal fluctuations select the ordering vectors $\langle\pi, \pi, \pi\rangle$, the yellow spots on the surface. The Brillouin zone boundary is shown as a light wireframe; the enclosing cube is a guide to the eye.

5. Problems studied by continuous-time quantum Monte Carlo method

Coulomb glass on the Bethe lattice
Phys. Rev. Research **4**, 023067 (2022) [arXiv:2009.02320]

The localization transition of disordered fermions in the presence of long-range Coulomb interactions represents challenging unsolved problems in modern condensed matter physics. Electrons form a glassy phase at strong interactions, which is indicated, among others, by an emerging pseudogap structure in the density of states. Thermal and quantum fluctuations melt the glass phase and induce a disordered Fermi liquid. Interestingly, a preformed pseudogap-like feature appears already in the Fermi liquid as a precursor of the quantum phase transition. We constructed a mean-field model for the quantum Coulomb glass and provided an accurate and extensive study of the model by self-consistent diagrammatic perturbation theory. Our calculations revealed a metallic glass phase at strong interactions, captured the emergence of a pseudo-gap structure at the Fermi energy, and provided the order parameter and the universal scaling properties deep inside the replica symmetry broken glassy phase. Combining self-consistent diagrammatic methods with numerically exact quantum Monte Carlo simulations, we determined the complete phase diagram and characterized the dynamics in the quantum liquid phase (see figure). We also applied the quantum Monte Carlo method for the glassy phase and obtained the pseudogap structure in the density of states. We found that the iterative perturbation theory gives results in excellent agreement with the numerically exact result proving its applicability for Coulomb glass models.



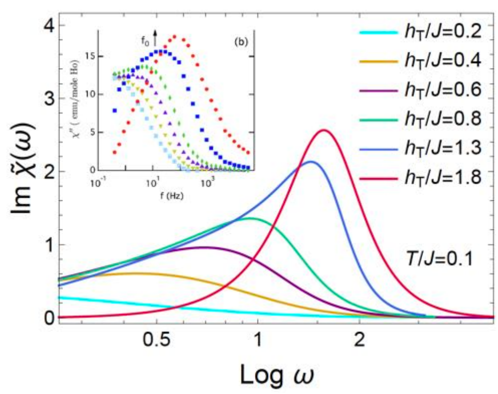
The density of states in the Fermi liquid phase displays a dip at the Fermi energy as a precursor of the spin glass quantum phase transition. We compare the results of iterative perturbation theory (IPT) with numerically exact continuous-time quantum Monte Carlo (CTQMC) simulations. The inset shows the temperature-interaction phase diagram with the Fermi liquid (FL) and Coulomb glassy (CG) phases.

Numerical studies of the zero-gap Kondo lattice model
Phys. Rev. B **100**, 214417 (2019) [arXiv:1906.05050]

We examined an indirect Padé approach as an analytic continuation for problems where quantum Monte Carlo methods calculate the Green's functions in imaginary time. We applied the Padé method to the conduction electron self-energy and then calculated the real-energy Green's functions from the analytically continued self-energy. This approach turned out to be very accurate and robust against statistical errors compared to the conventional direct analytic continuation of Green's functions, Next, we studied the Kondo lattice with vanishing conduction electron density of states at the Fermi level in infinite dimensions in the dynamical mean-field theory, where we used the indirect Padé approach to obtain accurate spectral functions. We found that a Kondo insulating ground state is realized only for Kondo couplings larger than a critical value, in contrast to the case of ordinary Kondo lattices. Moreover, we identify two energy scales from dynamic and thermodynamic properties which are shown to correspond to a direct and an indirect gap in a band hybridization picture.

Quantum Sherrington-Kirkpatrick spin-glass model by quantum Monte Carlo simulations
[A. Kiss, I. Lovas, C.P. Moca, G. Zaránd, in preparation]

The physics of disordered magnetic systems is a flourishing area of condensed matter physics. Among others, spin glass systems provide models for optimization problems in the field of biology, economics, society, or engineering. In spin glasses, the random distances between the spins lead to frustrations, since the signs and magnitudes of the interactions vary. As a consequence, the spins are randomly frozen in the ground state. We studied the infinite-range Ising spin glass model in a transverse field known as the quantum Sherrington-Kirkpatrick model. We explored the interplay of disorder and frustration with quantum effects using quantum Monte Carlo simulations. We provide the paramagnetic as well as the glassy solution in the whole temperature-transverse field plane and derived exact dynamics in the low-temperature spin glass state. The model is relevant for $LiHo_xY_{1-x}F_4$, a diluted derivative of the dipolar-coupled Ising ferromagnet $LiHoF_4$. We compared our results to a.c. susceptibility measurements and found a nice qualitative agreement, as shown in the figure**.**



Comparison between the numerically calculated (main panel) and experimentally observed (inset) dynamical susceptibility in the $LiHo_{0.198}Y_{0.802}F_4$ spin glass system (experimental data are from C. Ancona-Torres *et al*., Phys. Rev. Lett. **101**, 057201 (2008)).