Exotic phases of matter in quantum magnets

A tensor networks tale

Niesen, I.A.

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[245] *Private communication* with H.-Y. Lee.

Contributions to publications

Below is an overview of the contributions I made to the papers that form the basis of this thesis. According to custom, the authors appear in order of amount of material contributed: the first author being the main contributor, and the last serving as supervising (usually senior) scientist that oversees the overall progress of the paper in question.

All of the work listed below I performed under supervision of dr. P. Corboz.

[1] I. Niesen and P. Corboz
Emergent Haldane phase in the $S = 1$ *bilinear-biquadratic Heisenberg model on the square lattice*
Phys. Rev. B 95, 180404(R) (2017)

Discussed in **Chapter 3**

I performed the numerical work, except for the high-bond dimension Haldane simulations. I was also involved in the later stages of the writing process.

[2] I. Niesen and P. Corboz
A tensor network study of the complete ground state phase diagram of the spin-1 bilinear-biquadratic Heisenberg model on the square lattice
SciPost Phys. 3, 030 (2017)

Discussed in **Chapter 3**

I was responsible for all the numerical work, and also for writing the first draft of the article. The final version is a joint effort between me and my supervisor P. Corboz.

[3] I. Niesen, P. Corboz
Ground-state study of the spin-1 bilinear-biquadratic Heisenberg model on the triangular lattice using tensor networks
Phys. Rev. B 97, 245146 (2018)

Discussed in **Chapter 4**

Also for this paper, I carried out all the numerical work and wrote the first draft of the article. The final version of the paper came into being after a series of discussions with and feedback from P. Corboz.

[4] C. Boos, S.P.G. Crone, I. Niesen, P. Corboz, F. Mila and K.P. Schmidt
Plaquette phases in an extended Shastry-Sutherland model
in preparation

Discussed in **Chapter 5**

I ran initial simulations, and investigated the boundary-bond-dimension χ convergence. In addition, I supervised S.P.G. Crone with setting up his simulations, and provided feedback on his first draft. All writings in Chapter 5 come from my hand.

[5] S. Wessel, I. Niesen, J. Stapmanns, B. Normand, F. Mila, P. Corboz and A. Honecker
Thermodynamic properties of the Shastry-Sutherland model from quantum Monte Carlo simulations
arXiv:1808.02043 [cond-mat.str-el]

Discussed in **Chapter 6**

I executed all iPEPS simulations, and wrote a first draft for the iPEPS part of the paper. The final version of the iPEPS part of the paper is a collaborative effort of myself and P. Corboz.

Summary

Exotic Phases of Matter in Quantum Magnets

A Tensor Networks Tale

In many-body systems, the complex interplay between the particles that make up the system can lead to unexpected collective behavior, giving rise to *exotic phases of matter*. Well-known examples of such exotic phases are superfluids, which describe flow with zero viscosity; superconductors, which have zero electrical resistance and expel magnetic fields from within; and spin liquids: quantum many-body states that remain disordered down to zero temperature and can display topological order. Usually, these exotic phases involve the emergence of quantum mechanical effects at macroscopic length scales, which typically occurs in strongly correlated systems.

This thesis focuses on a subcategory of strongly correlated systems: insulating solids for which the dynamical particles—the electrons—are pinned to a background lattice, and the remaining degrees of freedom are their quantum mechanical spins. Such systems are called *quantum magnets*. Their constituent particles typically interact with Heisenberg-type interactions. Despite being described by relatively simple Hamiltonians, quantum magnets can exhibit very interesting physics, ranging from ordinary ferro and antiferromagnetism to nematic order and spin-liquid-like behavior.

The strong inter-particle interactions present in strongly correlated systems, and quantum magnets in particular, make them difficult to study, and force physicists to resort to numerical methods—especially for models of dimension higher than one. Due to the exponential scaling of the Hilbert space in the number of particles, however, brute-force numerical computations are doomed to fail for systems that consist of anywhere near the number of particles that real systems are made up of. Therefore, smart algorithms that allow for simulation of *large* strongly correlated systems are in great demand.

Based on statistical sampling, thus insensitive to the size of the many-body Hilbert space, quantum Monte Carlo (QMC) is one of the physicists' principal lines of attack. However, it turns out that a large class of strongly correlated systems—such as fermionic and frustrated systems—suffer from the *sign problem*, which makes QMC inviable. One of the main challenges of contemporary numerical many-body physics is to either come up with ingenious ways to circumvent the sign problem, or invent alternative methods.

This is where *tensor networks* come in. A many-body quantum state, which itself is one gigantic tensor, can be decomposed into a network made up of contractions of smaller local tensors. These local tensors carry a parameter, called the *bond dimension*, which determines how much entanglement can be captured by the tensor network state in question. Rather than performing numerical simulations on the entire Hilbert space, the simulations can be restricted to the much more manageable subset of fixed-bond-dimension tensor network states. If the needed level of accuracy requires it, the bond dimension can afterwards be extrapolated to infinity to obtain results that, in principle, are exact.

In this thesis several interesting quantum magnets are investigated by means of tensor networks. Specifically, we use *infinite projected entangled pair states* (iPEPS) to study two-dimensional quantum magnets directly in the thermodynamic limit; a necessary limit for the investigation of quantum phase transitions that separate different quantum phases.

Chapter 3 discusses the spin-1 bilinear-biquadratic Heisenberg (BBH) model on the square lattice. The BBH model is interesting experimentally because it contains the SU(3) Heisenberg model that can be realized using cold atoms. Moreover, being the most general lattice-translation, lattice-rotation and spin-rotation symmetric spin-1 system with nearest-neighbor interactions, the BBH Hamiltonian is also interesting from a theoretical point of view. Besides the known magnetic and nematic phases, our tensor network study of the square lattice BBH model demonstrates the occurrence of two additional phases that had been overlooked by previous studies: a partially magnetic partially nematic phase, and a paramagnetic phase that preserves spin-rotation and lattice-translation symmetries but breaks lattice-rotation symmetry. By studying the anisotropic square lattice BBH model, we continue to show that this paramagnetic phase can be adiabatically connected to the well-known Haldane phase of decoupled spin-1 chains.

Motivated by the unusual and not-well-understood behavior of the triangular lattice quantum magnets $NiGa_2S_4$ and $Ba_3NiSb_2O_9$, we then proceed to investigate the triangular lattice BBH model in Chapter 4. Our iPEPS simulations reproduce both the ferroquadrupolar and antiferroquadrupolar phases that were suggested to be realized in $NiGa_2S_4$. Interestingly, both the partially magnetic partially nematic phase as well as the extended Haldane phase found on the square lattice turn out to be absent from the triangular lattice phase diagram. However, our study does show that the triangular lattice BBH model is susceptible to lattice distortions, and that even in the absence of the biquadratic coupling, the system on the triangular lattice undergoes a transition to the Haldane phase at about

80% anisotropy.

Chapter 5 focuses on a different quantum magnet, the *Shastry-Sutherland* (SSL) model. This model has been studied extensively because (i) it is one of the few two-dimensional magnets for which an exact eigenstate—made up of a product dimer singlets—is known, and (ii) it is realized experimentally in $SrCu_2(BO_3)_2$. The dimer eigenstate is actually the ground state of the SSL model for weak inter-dimer coupling. For strong inter-dimer coupling, the ground state is a Néel ordered square lattice antiferromagnet (AFM). There has been ongoing debate on what happens in between the dimer and AFM phases, but most works agree that there exists an intermediate plaquette phase. However, the nature of the plaquette phase has been questioned by a recent experimental paper that examined $SrCu_2(BO_3)_2$ under external pressure, and argued in favor of the so-called *full plaquette phase*; this in contrast to the conclusion stated in other recent theoretical papers that claim that the *empty* rather than the full plaquette phase occurs in between the dimer and AFM phases. By directly comparing the energies of both plaquette states, we show that the intermediate phase is the empty plaquette phase. However, a bias in the Hamiltonian towards the full plaquette phase of only a few percent makes the full plaquette state the lowest energy state, which shows that the SSL model is susceptible to anisotropies. It is not unlikely that subjecting $SrCu_2(BO_3)_2$ to external pressure distorts the lattice non-uniformly, making the full plaquette state energetically favorable over the empty plaquette one.

Finally, Chapter 6 examines an extension of the SSL model (which interpolates between the ordinary SSL and the fully-frustrated bilayer models) in the context of the sign problem in QMC. It turns out that, when expressed in the dimer basis, the sign disappears at low temperatures. This has to do with the fact that the dimer ground state of the SSL model is also the ground state of an artificial sign free model obtained by making all off-diagonal elements of the Hamiltonian non-positive (in the dimer basis). A comparison with our iPEPS study shows that, as expected, the regime wherein the sign problem disappears is contained within the dimer phase of the extended SSL model. Using QMC, the magnetic susceptibility and specific heat are computed for the SSL model. The above demonstrates that knowledge of an exact ground state can possibly be used to accurately compute thermodynamic quantities of frustrated systems by means of QMC.

Samenvatting

Exotische Fases van Materie in Kwantummagneten

Een tensornetwerkverhaal

In meerdeeltjessystemen kan de complexe wisselwerking tussen de deeltjes die het systeem vormen, leiden tot onverwacht collectief gedrag, wat aanleiding kan geven tot *exotische fases van materie*. Bekende voorbeelden van zulke fases zijn supervloeistoffen, die stroming zonder viscositeit beschrijven; supergeleiders, die geen elektrische weerstand ondervinden en magnetische velden van binnenuit verdrijven; en spinvloeistoffen: meerdeeltjes kwantumtoestanden die ongeordend blijven tot nul graden en topologische orde kunnen vertonen. Doorgaans gaan dit soort exotische fases gepaard met de verschijning van kwantummechanische effecten op macroscopische lengteschalen, hetgeen typisch voorkomt in sterk-gecorreleerde systemen.

Deze thesis richt zich op een subcategorie van sterk-gecorreleerde systemen: isolerende vaste stoffen waarvoor de bewegende deeltjes—de elektronen—vastgespeld zitten op een achtergrondrooster, en de enige overgebleven vrijheidsgraden worden gegeven door de kwantummechanische spins van de elektronen. Zulk soort systemen worden *kwantummagneten* genoemd. De deeltjes die deze systemen vormen gaan doorgaans Heisenberg-achtige interacties met elkaar aan. Ondanks het feit dat zij door relatief eenvoudige Hamiltonianen worden beschreven, kunnen kwantummagneten zeer interessante fysica tonen, variërend van ordinair ferro- en antiferromagnetisme tot nematisch en spinvloeistofachtig gedrag.

De sterke interdeeltjesinteracties die aanwezig zijn in sterk-gecorreleerde systemen, en in kwantummagneten in het bijzonder, maken dat dit soort systemen lastig te bestuderen zijn, hetgeen natuurkundigen dwingt toevlucht te nemen tot numerieke methodes—vooral voor systemen in dimensies hoger dan één. Echter, wegens de exponentiële schaling van de Hilbertruimte in het aantal deeltjes, zijn rechttoe rechtaan simulaties gedoemd om te falen voor systemen die bestaan uit een aantal deeltjes dat ook maar enigszins in de buurt komt van het aantal deeltjes waar een werkelijk fysische systeem uit bestaat. Derhalve is er grote vraag naar slimme algoritmes die het mogelijk maken om *grote* sterk-gecorreleerde systemen te simuleren.

Kwantum Monte Carlo (KMC), een methode die gebaseerd is op statistische bemonstering en daarom ongevoelig is voor de grootte van de meerdeeltjes Hilbertruimte, is een van de primaire onderzoeksmethodes die natuurkundigen tot hun beschikking hebben. Helaas blijkt een grote klasse van sterk-gecorreleerde systemen—zoals fermionische en gefrustreerde systemen—onderhevig te zijn aan het zogeheten *tekenprobleem*, hetgeen KMC ontoereikend maakt. Een van de grote uitdagingen van de hedendaagse numerieke meerdeeltjesfysica bestaat uit ofwel het bedenken van ingenieuze wijzen om het tekenprobleem te omzeilen, ofwel het ontwikkelen van alternatieve methoden.

Dit is waar tensornetwerken hun entree maken. Een meerdeeltjestoestand, op zich een gigantische tensor, kan ontleed worden in een netwerk gevormd door contracties van kleinere lokale tensoren. Deze lokale tensoren brengen een parameter—de *bonddimensie*—met zich mee, die bepaalt hoeveel verstrengeling er bevat kan worden in de tensornetwerktoestand in kwestie. In plaats van simulaties uit te voeren op de gehele Hilbertruimte, kunnen simulaties beperkt worden tot de beheersbaardere deelverzameling van vaste-bonddimensie tensornetwerktoestanden. Indien nodig, dan is het mogelijk om de bonddimensie naar oneindig te extrapoleren en zo resultaten te verkrijgen die in principe exact zijn.

In deze thesis worden verscheidene interessante kwantummagneten onderzocht door middel van tensornetwerken. Preciezer gezegd gebruiken wij *oneindige geprojecteerde verstrengelde paartoestanden* (oGVPT) om tweedimensionale kwantummagneten te bestuderen in de thermodynamische limiet; een limiet die noodzakelijk is voor het onderzoeken van kwantumfasetransities die plaatsvinden tussen twee verschillende kwantumfases.

Het onderwerp van discussie van Hoofdstuk 3 is het spin-1 bilineair-bikwadratisch-Heisenbergmodel (BBHM) op het vierkant rooster. Het BBHM is interessant vanuit experimenteel oogpunt omdat het het SU(3)-Heisenbergmodel bevat dat gerealiseerd kan worden middels koude atomen. Bovendien is het BBHM, als het meest algemene roostertranslatie-, roosterrotatie- en spinrotatiesymmetrische spin-1 systeem met naaste-buurinteracties, interessant vanuit theoretisch perspectief. Onze tensornetwerkstudie van het BBHM op het vierkant rooster laat zien dat er, naast de bekende magnetische en nematische fases, twee bijkomende fases voorkomen die door eerdere studies over het hoofd gezien zijn: een deels-magnetische deelsnematische fase, en een paramagnetische fase die spinrotatie- en roostertranslatiesymmetrie bewaart maar roosterrotatiesymmetrie breekt. Door het anisotropische BBHM te bestuderen tonen wij vervolgens aan dat deze paramagnetische fase adiabatisch verbonden kan worden met de bekende Haldanefase van ontkoppelde spin-1 ketens.

Gemotiveerd door het ongebruikelijke en niet-volledig begrepen gedrag van de driehoekig rooster kwantummagneten $NiGa_2S_4$ en $Ba_3NiSb_2O_9$, zetten wij ons werk voort met een vervolgstudie van het BBHM op het driehoekig rooster in Hoofdstuk 4. Onze oGVPT-simulaties reproduceren zowel de ferroquadrupolaire als ook de antiferroquadrupolaire fases waarvoor gesuggereerd is dat zij gerealiseerd worden in $NiGa_2S_4$. Opmerkelijk is het feit dat zowel de op het vierkant rooster aangetroffen deels-magnetische deels-nematische alsmede de Haldanefase afwezig blijken te zijn in het fasediagram van het BBHM op het driehoekig rooster. Echter, onze studie toont wel aan dat het BBHM op het driehoekig rooster fijngevoelig is voor roostervervormingen, en dat, zelfs in afwezigheid van de bikwadratische koppeling, het systeem op het driehoekig rooster een fasetransitie ondergaat naar de Haldanefase bij ongeveer 80% anisotropie.

Hoofdstuk 5 beschouwt een ander type kwantummagneet: het *Shastry-Sutherland-model* (SSM). Dit model is reeds uitgebreid bestudeerd omdat (i) het een van de weinige tweedimensionale magneten is waarvoor een exacte eigenstoestand, bestaande uit een produkt van dimeer singlets, bekend is, en (ii) het experimenteel gerealiseerd wordt in $SrCu_2(BO_3)_2$. De dimeereigentoestand is zelfs de grondtoestand van het SSM voor zwakke interdimeerkoppeling. Voor sterke interdimeerkoppeling is de grondtoestand een Neél-geordende vierkant-rooster antiferromagneet (AFM). Wat er gebeurt tussen de dimeer- en AFM-fases is onderwerp van debat, maar de meeste studies zijn het erover eens dat er een tussenliggende plaquettefase bestaat. Echter, de geaardheid van deze plaquettefase is in twijfel getrokken in een recent experimenteel artikel dat $SrCu_2(BO_3)_2$ onderwierp aan externe druk, en pleit voor het bestaan van de zogeheten *volle plaquettefase*; dit in tegenstelling tot de conclusie van de overige recente artikelen die beargumenteren dat de *lege* in plaats van de volle plaquettefase zich voordoet te midden van de dimeer- en AFM-fases. Door de energieën van beide plaquettetoestanden direkt te vergelijken, demonstreren wij dat de tussenliggende fase de lege plaquettefase is. Doch, een afwijking in de Hamiltoniaan ten gunste van de volle plaquettefase van slechts een paar procent maakt dat de volle plaquettetoestand de laagste energietoestand wordt, hetgeen demonstreert dat het SSM vatbaar is voor anisotropiën. Het is niet ondenkbaar dat het blootstellen van $SrCu_2(BO_3)_2$ aan externe druk het rooster vervormt op dusdanige niet-uniforme wijze, dat de volle plaquettetoestand energetisch bevoordeeld wordt ten opzichte van de lege plaquettetoestand.

Tot slot onderzoeken wij in Hoofdstuk 6 een extensie van het SSM (dat tussen het ordinaire SSM en het volledig-gefrustreerde dubbellaags model interpoleert) in de context van het tekenprobleem in KMC. Het blijkt dat, wanneer er wordt gewerkt in de dimeerbasis, het teken verdwijnt bij lage temperaturen. Dit gegeven is gerelateerd aan het feit dat de dimeergrondtoestand van het SSM ook de grondtoestand is van een artificieel tekenvrij model verkregen door alle niet-diagonaalelementen van de Hamiltoniaan niet-positief te maken (in de dimeerbasis). Vergelijking met onze oGVPT-studie laat zien dat, zoals verwacht, het regime waarin het tekenprobleem verdwijnt, onderdeel uitmaakt van de dimeerfase van het uitgebreide SSM. Gebruikmakend van KMC berekenen wij de magnetische susceptibiliteit en de warmtecapaciteit van het SSM. Het bovenstaande demonstreert dat kennis van een exacte grondtoestand mogelijkerwijs benut kan worden om thermodynamische grootheden van gefrustreerde systemen nauwkeurig te berekenen door middel van KMC.

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והרבה אהבה ותודה לכל המשפחה, בשביל כל מה שעשיתם בשבילי לאורך השנים.

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