論文の内容の要旨　Abstract of Dissertation

Geometrically Constructed Markov Chain Monte Carlo Study of Quantum Spin-phonon Complex Systems
(幾何学的構成マルコフ連鎖モンテカルロ法による量子スピンフォノン複合系の研究)

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1. Background and Purpose

Frustrated quantum spin systems contain rich physical structures of nontrivial quantum states and phase transitions, and it is very important to elucidate the mechanism as a grand challenge problem in the condensed matter physics and the material design. The origin of frustration can be a geometric structure or an interaction with other degrees of freedom. Among them, the spin-lattice interaction has a large contribution for determination of effective spin-spin interaction. As a system dominated by the spin-lattice interaction, a spin-Peierls system has caught the attention for a long time. When the decrease in energy by dimerization that makes spin singlet pairs
exceeds the increase by lattice distortion, the spin-Peierls system turns into the dimer phase, which is called the spin-Peierls transition. The analysis of this system and transition will bring understanding of the role of lattice degree of freedom in condensed materials.

Two limiting cases have been considered in the spin-Peierls system: the adiabatic limit and the antiadiabatic limit. The former case is realized in some organic materials experimentally; for example, the compound TTF-CuBDT was found as the first spin-Peierls material in 1975. The low-energy physics is described by a S=1/2 one-dimensional Heisenberg chain coupled with the lattice degree of freedom in harmonic potential. Cross and Fisher analyzed this model by using the Abelian bosonization method combined with the random phase approximation in the adiabatic limit, where the quantum nature of lattice is ignored. Their theory seems effective to the organic materials because a soft phonon mode that is one of consequences of their theory was observed in experiments.

The Cross-Fisher theory is considered to be, however, not valid to the compound CuGeO$_3$ that was discovered as the first inorganic spin-Peierls material in 1993. It is because that the energy scale of phonon is the same order with that of spin, which is away from the adiabatic region, and no soft phonon was observed experimentally. This discovery triggered many theoretical approaches from the antiadiabatic limit: the perturbation expansion, the flow equation method, the unitary transformation, etc. According to these approaches, tracing out the phonon degree of freedom results in producing an effective spin frustration. Then, the quantum spins construct the dimer by this frustration effect as the famous Majumdar-Ghosh point. Interestingly, the phase diagram and the universality class of transitions are totally different between the adiabatic and antiadiabatic limits. Not only for understanding this qualitative difference but also for explaining the behavior of realistic materials away from the two limits, such as CuGeO$_3$ and MEM(TCNQ)$_2$, it is important to analyze the crossover and connection

between them. A precise calculation for the intermediate region, however, has been difficult because of the complexity of the system. On these backgrounds, the purpose of this thesis are to invent a novel method that can treat the spin-Peierls system correctly and to elucidate the mechanism of the quantum phase transitions triggered by the quantum nature of lattice degree of freedom.

2. Developments of New Methods

We present three novel methods in this thesis. The first is a generic improvement of sampling efficiency in the Markov chain Monte Carlo (MCMC) method. The MCMC method is a generic tool for investigating many kinds of systems with multiple degrees of freedom. For the method to work effectively, we must consider following three key matters: the choice of ensemble, the selection of candidate states, and the optimization of transition kernel. For the first, the extended ensemble methods, such as the multicanonical method, have been applied to many hardly-sampled systems. For the second, the cluster algorithms, such as the Swendsen-Wang algorithm, can perform a global update with low or zero rejection rate. For the third, the Metropolis algorithm or the heat bath algorithm (Gibbs sampler) is widely used in practical simulations. However, these conventional methods are not optimal. In the meantime, since the invention by Metropolis and the co-workers in 1953, the MCMC method has evolved within the paradigm of the detailed balance, namely reversibility. The detailed balance is, however, not necessary condition. Instead of solving the usual algebraic problem of the detailed balance equation, we rewrite the needed task as a geometric allocation problem and always find not only a reversible solution but also an irreversible kernel with minimized rejection rate. Our approach is a new kind of optimization method that is particularly different from conventional schemes using derivatives of a cost function. As an important point, there is no additional CPU time cost in our method. This algorithm is the first versatile method that is free from the detailed balance. By applying our algorithms for the 8-state Potts model, the autocorrelation time of the single spin flip update gets more than 12 times as short as that by the Metropolis algorithm. In addition, the correlation time for the quantum Heisenberg chain with magnetic field becomes more than 100 times as short as that by the heat bath algorithm. Moreover, our algorithms can be generally extended to also continuous variables. Thus, the methods presented in this thesis should replace the conventional update procedures in all MCMC simulations. This optimization is significantly effective to also following our update method for the spin-Peierls model.

The second is the development of new quantum Monte Carlo (QMC) method for particle number nonconserving systems, such as the spin-Peierls model written by the second quantization. The QMC method that is based on the worldline representation have been applied to a wide variety of quantum spin, bosonic and fermionic systems. In recent years, the method has passed through two turning points of key improvements. One is the elimination of the decomposition error of imaginary time; Beard et al. showed the formalism where it becomes possible to simulate quantum systems directly in continuous time. The other is the invention of nonlocal worldline update methods, such as the loop algorithm and the worm algorithm. Later,

Syljuasen et al. extended the worm update to a more efficient procedure, which has become the standard method now, which is called also the directed-loop algorithm. In the meanwhile, we need to treat a nonconserved particle when a good quantum number and basis set are difficult to find. In our spin-Peierls model, soft-core bosons (phonons) are not conserved by the Hamiltonian. The conventional worm (directed-loop) algorithm is, however, not applicable to nonconserved particles because of the ergodicity problem. Thus, we have extended the worm update so that it becomes possible to calculate such models. Moreover, we have successfully removed the worm bounce process, which has been a bottleneck of the method, by combining our optimized kernel as we mentioned above. By these improvements, we can treat general conserved/nonconserved quantum spins and bosons without any approximation (typically, decomposition error or occupation number cutoff of soft-core bosons). As a result, a precise Monte Carlo analysis of large-scale spin-Peierls systems has become feasible for
the first time.

The third is the invention of an estimation method for precise energy gap and of the level spectroscopy by the QMC method. Using our extended worm algorithm, we have found that the Kosterlitz-Thouless (KT) type phase transition occurs in the one dimensional spin-Peierls systems. For the KT type transition, the conventional finite-size scaling method, where the correlation length is assumed to diverge in a power-law form, fails to catch the critical point. In the meanwhile, the level spectroscopy, where a transition point and an universality class can be determined by using excitation gaps even for the KT transition, has been applied to many one-dimensional quantum systems. In this thesis, we have combined this method with the QMC technique and successfully applied to the spin-Peierls systems. For the combination, we need to calculate many excitation gaps precisely. The problem of gap estimation in the QMC method is, however, ill-posed as the inverse Laplace transformation; the conventional estimator of gap has not only statistical error but also systematic error. In order to get a precise gap, we have invented new estimator sequence that converges to a true gap value. Moreover, we have formulated new calculation procedures to measure the excitation gap of the S=0 singlet and the S=1 S^z=+1,-1 doublet, although only the gap of S=1 S^z=0 is available in the conventional approach. Thus, we can use the level spectroscopy analysis with precise gap estimations.

3. Quantum Phase Transition of Spin-Peierls Systems

We have applied above novel methods to the XXZ spin-Peierls chain and two dimensional system coupled to the quantum phonon of optical mode. In the one-dimensional XXX (Heisenberg type) spin-Peierls system, an infinitesimal spin-lattice coupling drives the ground state into the dimer state in the adiabatic limit, as the Cross-Fisher theory. On the other hand, the quantum phase transition from the Tomonaga-Luttinger liquid phase to the dimer phase occurs at a finite coupling parameter when the quantum nature of lattice is taken into account. The phase transition has been believed to be a KT type from the analogy with the frustrated J1-J2 model. The transition point has been estimated by using some analytical method, such as the flow equation method or the unitary transformation. These estimated points, however, have differed

considerably between each others. We have determined the critical point much more precisely than ever by applying the developed gap-estimation method with the level spectroscopy. We have determined also the universality class, and elucidated the phase diagram of the one dimensional XXZ spin-Peierls system for the first time. For the XY-like anisotropy, the phase transition between the Tomonaga-Luttinger liquid phase and the dimmer phase occurs and the low-energy physics on the transition line is governed by the SU(2) k=1 Wess-Zumino-Witten model. For the Ising-like model, on the other hand, the transition line of the Gaussian universality class appears between the Neel phase and the dimmer phase. The whole phase diagram is qualitatively consistent to the sine-Gordon model. This correspondence is reasonable because the original spin-Peierls model can be renormalized to the frustrated J1-J2 model and further to the sine-Gordon model, according to the antiadiabatic approach. However, we have confirmed the aspect of this phase diagram still remains even in the adiabatic parameter region. This correspondence is not trivial because it implies the universality class instantaneously changes no matter how small the quantum fluctuation of lattice degree of freedom is. This finding manifests the essence of the quantum nature of lattice in the spin-Peierls systems.

We also investigated a two dimensional spin-Peierls system. For higher dimensions than one, there have been only few researches so far because the bosonization method on which most analytical approaches have relied cannot be effective. We have considered an interesting two dimensional system where the spin-Peierls chains are connected only by phonon interchain interaction without interchain spin interaction. For a strong spin-phonon coupling region of this model, a finite-temperature phase transition from the disorder phase to the dimer phase occurs. On the other hand, for a weak spin-phonon coupling region, we have found a interesting two-dimensional gapless phase that has a similar property with the one-dimensional case. This gapless excitation certainly stems from the effective spin frustration beyond the mean-field treatment. This phase is the first discovery of the two dimensional gapless quantum liquid phase by the worldline Monte Carlo method.

4. Conclusion

We have developed the three novel methods for investigating the spin-Peierls system correctly. These methods are generally applicable to other physical and statistical problems. We have applied our methods to the one and two dimensional large-scale spin-Peierls systems. As a result, we have elucidated the phase diagram of the one-dimensional XXZ spin-Peierls model and found the interesting quantum liquid phase in the two-dimensional system. These analyses are the first precise calculations of the quantum criticality not only for the spin-Peierls model but also for effectively frustrated quantum spin systems by the worldline Monte Carlo method.