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Author(s): T. E. Booth, R. A. Forster, J. A.
Carlson, M. A. Gomez, J. E. Gubernatis,
N. Hatano, L. R. Pratt, J. R.
Quagliano, J. C. Scovel, R. N. Silver,
S. Zhang, C. R. Doering, J. D. Doll, N.
Kawashima

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Simulation Methods for Advanced Scientific Computing

Thomas E. Booth, Joseph A. Carlson, R. Arthur Forster, Maria A. Gomez,
James E. Gubernatis*, Naomichi Hatano, Laurence R. Pratt, John R. Quagliano,
James. C. Scovel, Richard N. Silver, and Shiwei Zhang
Los Alamos National Laboratory

Charles R. Doering
University of Michigan
Jimmie D. Doll
Brown University

Naoki Kawashima
Toho University, Japan

Abstract

This is the final report of a three-year, Laboratory Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). The objective of the project was to create effective new algorithms for solving N-body problems by computer simulation. We concentrated on developing advanced classical and quantum Monte Carlo techniques. For simulations of phase transitions in classical systems, we produced a framework generalizing the famous Swendsen-Wang cluster algorithms for Ising and Potts models. For spin-glass-like problems, we demonstrated the effectiveness of an extension of the multicanonical method for the two-dimensional, random bond Ising model. For quantum mechanical systems, we generated a new method to compute the ground-state energy of systems of interacting electrons. We also improved methods to compute excited states when the diffusion quantum Monte Carlo method is used and to compute longer time dynamics when the stationary phase quantum Monte Carlo method is used.

Background and Research Objectives

The rapid increase in raw computational power has created exciting opportunities for scientific computation. The complexity and realism of the problems that can be simulated promise to greatly advance many scientific and technological fields. Accordingly, scientific computing is enjoying considerable worldwide popularity. Yet many challenging scientific problems remain unsolved due to inadequate algorithms, especially algorithms that are suitable for massively parallel architectures. In this project our objective was to create effective new algorithms for solving these problems, giving the Laboratory the initiative in key technical areas by combining the talents and efforts of our

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*Principal Investigator, e-mail: jg@lanl.gov

physical scientists and applied mathematicians. We concentrated on developing advanced N-body simulation techniques, focusing on classical and quantum Monte Carlo techniques.

Specifically, we tackled two problems intrinsic to classical N-body simulation methods. One addressed improving cluster algorithms for studying phase transitions. The other addressed improving methods for systems with highly multimodal distributions of energy like the spin glass problem. One of the most significant breakthroughs in computer simulations of phase transitions was the Swendsen-Wang cluster algorithm. It completely changed the way computer simulations of phase transition for Ising-Potts-like models are done and its two to four magnitude reduction in computer time broadened the potential application of such activities. We sought to extend the cluster algorithm concept to systems other than Ising-Potts models, and we were successful for a variety of lattice models.

In many respects the "spin glass" problem is the paradigm bottleneck for N-body simulations. Here "spin glass" is just a rubric for problems with both disorder and frustration. Frustration commonly occurs in N-body simulations; for example, it is an intrinsic feature of simulations of vortex motion and pinning in superconductors. Understanding pinning is essential to understanding these materials, and thus to designing better ones. Roughly speaking, frustration is the inability of many local regions of particles to cooperate simultaneously to obtain a lower energy configuration. When it is present, computer simulations become several orders of magnitude more difficult than normal, and effective algorithms exist for only a few special cases. Our approach was to implement and test the effectiveness of a recently proposed multicanonical algorithm for the random spin problems and to develop extensions of this method.

We also tackled three problems intrinsic to quantum N-body simulation methods: computing the ground-state properties of systems of interacting electrons, computing excited states of quantum systems, and computing quantum dynamics. The first problem is intensified by the infamous fermion sign problem. This problem causes the variance of computed results to grow with system size and has prevented some quantum simulations from studying large enough systems to determine convincingly the phase of the ground state. We developed a new variational approach to the problem that appears to be highly accurate.

The problem of excited states is related to the problem of determining the ground-state, but here whether the system is fermionic or not is irrelevant. Quantum Monte Carlo methods have had problems determining multiple excited state of a system with accuracy comparable to the determination of the ground-state energy. We revisited the problem in

the context of the diffusion quantum Monte Carlo method and extended existing methods into one that appears more accurate.

The dynamics of physical processes are often more important than the equilibrium or ground-state properties. For classical problems, the molecular dynamics technique is a well-established procedure for obtaining such information for many processes. For quantum problems, the dynamical techniques are not as well established because of intrinsic difficulties in doing quantum simulation in real time and in inverting imaginary-time simulations to real time. We studied extensions of real-time methods.

Importance to LANL's Science and Technology Base and National R&D Needs

The Nation's drive to newer technologies often requires understanding phenomena at the particle level. For example, predicting the macroscopic properties of novel materials requires an understanding of the microscopic, or particle, level behavior of those materials. Here, the difficulty in using analytic methods mandates the use of N-body methods. We have identified several impediments limiting the utility of N-body methods above, and we seek to overcome them by improving existing algorithms and creating new ones. Many different disciplines share these algorithms, so advancing one class of algorithms will generally impact several disciplines.

Tomorrow's computers will enable the investigation of even more complex physical problems. In some cases, N-body simulations of classical systems are already beginning to produce macroscopic descriptions that rival those produced by continuous simulations, so these two approaches are beginning to complement each other. In the near future, we believe that new computer architectures will enable N-body simulations to be competitive with continuum simulations, and we will be able to use both approaches to enhance our understanding of complex physical problems.

Scientific Approach and Accomplishments

Different disciplines like astrophysics, biophysics, chemistry, condensed matter physics, materials science, nuclear physics, and high energy physics generally have different applications, but all use one of three basic N-body methods: molecular dynamics, Monte Carlo, and stochastic differential equations (Langevin methods), and consequently all share the difficulties and limitations of these methods.

The Monte Carlo method is the most flexible of the three methods and has been used for a variety of classical and quantum simulations. For equilibrium problems, the Metropolis algorithm ensures that configurations are produced with Boltzmann weights, but does not specify how to do this efficiently. For some physical systems, like those near

a phase transition or those with frustration, the simulations become extremely inefficient. Recently, cluster methods were developed by Swendsen and Wang for Ising and Potts models, which greatly reduced the simulation time for phase transitions, basically by replacing traditional local configuration changes in the Metropolis algorithm by large-scale coherent ones. More recently, we proposed a general prescription for developing cluster algorithms of lattice models (1).

Our general framework for constructing cluster algorithms had a significant "spin-off." With it, a preliminary start was made at impacting a broader class of problems areas. With new algorithms and perspectives, we already impacted significantly at least one technique, the world-line method, which is computationally intense even away from a phase transition, and have also impacted two quantum problems areas, the one-dimensional Hubbard (1) and quantum spin models (2,3,4).

As the model of a spin glass we chose the random-bond Ising model. For this model the major unresolved issue is the definition of the zero-temperature (ground) state in three dimensions. Even when the temperature is lowered below the glass transition, simulations experience a type of critical slowing down due to the presence of many low-lying metastable states. The issue is whether or not the ground state is infinitely or doubly degenerate. We applied several Monte Carlo approaches to the problem. First we used the Swendsen-Wang replica method. While highly successful in two dimensions this method proved to be inefficient in three. We next tried the multicanonical method. This method is very effective for problems with multi-modal distribution of energy-like bimodal distribution encountered near a first-order phase transition and the more complicated distribution encountered in polymer systems. Its effectiveness for low temperature spin glasses was less certain. The method seeks to adaptively determine the energy distribution during the course of the simulation. While it is better than more direct Monte Carlo methods, we sought to further improve the efficiency by determining the distribution of the order parameter. This modification did not make any significant gain. We then determined the joint distribution of the energy and the order parameter. In two dimensions a significant improvement was made. We are now implementing the method in three dimensions and preliminary results are encouraging.

Research activities and collaborations initiated or associated with the project produced a landmark development for ground-state algorithms for quantum Monte Carlo simulations (5,6). Our newly developed method now provides the first feasible method for simulating quantum systems of strongly interacting electrons of sufficient size and accuracy to test where models such as the two-dimensional Hubbard model exhibit superconductivity. The new method eliminated the fermion sign problem by trading a sign-

problem-plagued, exact method for a sign-problem-free, highly accurate variational method.

With the new method, which we call the Constrained-Path Monte Carlo, we performed simulations on two of the most studied models for high temperature superconductivity, the one- and three-band Hubbard models (7). With the superior numerical accuracy of the method, we were able to pull out of the statistical noise superconducting pairing correlations. The correlations however were also demonstrated to disappear as the interactions strength was increased and also to apparently disappear as the system size was increased. The latter finding in particular should be discouraging for those studying these models as paradigms for high-temperature superconductivity: it is inconsistent with the presence of off-diagonal, long-range superconducting order.

The importance of chemical reactions in liquid water has sparked significant interest in understanding proton transfer phenomena in water. The quantum mechanical nature of proton transfer in liquid water necessitates the development of new quantum mechanical techniques. If the density matrix of a strongly quantum mechanical system were known, it could be used to calculate various equilibrium properties including the effects of quantum mechanics. The density matrix can be constructed from the excited states of the system that are relevant to the temperature range desired. Even if excited states were only known for a smaller subsystem that contained most of the quantum mechanical effects, a density matrix for the whole system can be constructed from a product of the density matrix for the subsystem times a Boltzmann factor with a "Feynmann-Hibbs potential".

Diffusion Monte Carlo algorithms have been used extensively to calculate ground state properties. The calculation of excited states, however, is significantly more difficult due to difficulties in specifying the location of nodes. Recently, Bernu, Ceperley, and Lester developed Correlation Function Quantum Monte Carlo, a method capable of finding multiple excited state energies and wavefunctions simultaneously. This method requires reasonable initial guesses for the ground and excited state wavefunctions. We have developed a scheme for obtaining initial wave functions using the short time (high temperature) approximation to the density matrix. This method yields reasonable guiding functions. As a test case, we are calculating the ground and a few excited states for D_3O^+ using the OSS potential.

Most quantum Monte Carlo simulations are performed in imaginary time; that is, real (physical) time is analytically continued to the imaginary axis. This converts a wave propagation problem into a diffusion one that is easier to simulate by a Monte Carlo technique. Taking the results of the simulation from imaginary time to real time can be very

difficult. However, real-time simulation methods exist. The challenge is to extend them so that they produce reliable results other than at short-times.

The most successful real-time quantum Monte Carlo method is the stationary-phase method. In this method the Monte Carlo sampling is biased to sample worldlines that on the average keep the phase of the exponential of the action constant. An extension of the method was made that can be most simply described as a reweighting of this sampling. Initial results on a toy problem demonstrated significant computational gains. Extensions to more physically representative models is in progress.

Part of the project's activities was its co-sponsorship of a visitor and a workshop program. In both programs, Laboratory personnel were placed in contact with leading experts in the fields, disseminating information on the latest developments on methods and their applications.

A three-day workshop, titled "Algorithms for Computer Simulations of N-Body Problems," was held in January, 1997. Thirteen external and three Los Alamos speakers summarized the state of the art in numerical algorithms in key application areas for simulations of both classical and quantum mechanical systems. Topics included the newest algorithms for time interaction of molecular dynamics and stochastic differential equations, the latest developments in extracting real-time information from quantum mechanical simulations, the newest breakthrough in quantum ground-state calculations, and the latest developments in nonequilibrium simulations for surface catalysis and chemical reactions. The workshops had nearly 80 registrants, of which 60 were from the Laboratory.

One consequence of the workshop was the interest it developed among the carefully chosen external participants about visiting the Los Alamos Center for Nonlinear Studies (CNLS), sending graduate students here, and exploring opportunities to place students in post-doctoral positions. One participant (J. Doll) decided to spend a sabbatical half-year at the Laboratory; four others revisited. Additionally, two graduate students visited as a preliminary step towards possibly doing part of their doctoral thesis research at the Laboratory.

A second workshop on "Adaptive Monte Carlo Methods" was co-sponsored with the Applied Theoretical and Computational Physics (X) Division. Here the emphasis was on nonequilibrium problems, in particular on the problem of radiation transport. The meeting provided a forum for internationally based leaders in the field to review state-of-the-art methods in variance reduction techniques for transport Monte Carlo methods. It also generated widespread interest in recent developments in X Division on variance reduction methods that adaptively learn how to produce a zero variance solution. The

meeting was so successful that before it ended plans were put into place to hold a second one in 1998.

A third workshop on "High Performance Computing In Chemistry" was co-sponsored with the Chemical Science and Technology (CST) Division. This was a one day, internal event designed to bring scientific problems in analytical chemistry that CST Division is facing to a broader Laboratory audience traditionally more skilled in mathematical modeling and large scale usage of computers. The workshop participants came from five different technical divisions at the Laboratory. Several points of contacts between CST staff and other Laboratory staff developed which have led to several collaborations and joint proposals. A second workshop will be held at the CNLS before the end of 1997.

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