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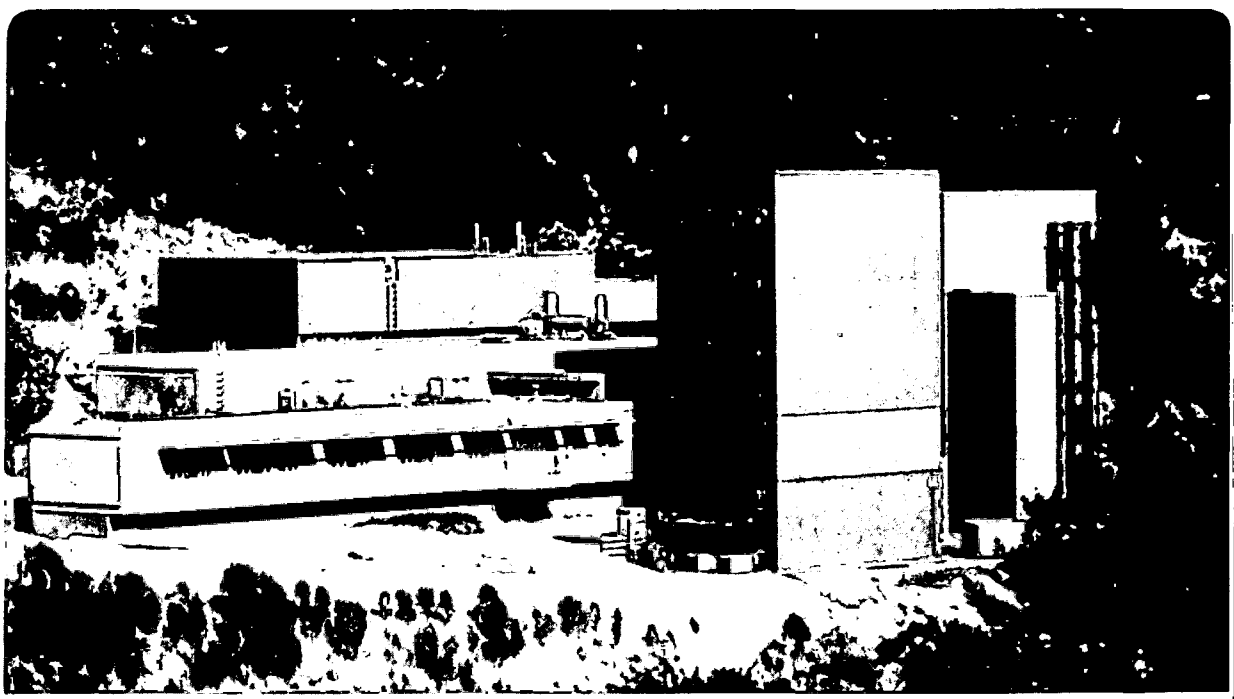

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Monte Carlo Simulation of Oxygen Ordering in the High-Temperature Superconductor $YBa_2Cu_3O_z$ *

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Abstract

The development of Monte Carlo single instruction multiple data (SIMD) parallel algorithms for the study of oxygen ordering in the basal plane of $YBa_2Cu_3O_z$ from first principles is described. In particular, implementation of the Ising model with short-range pair interactions on the SIMD architecture MasPar MP-1 (DEC mpp-12000) series of massively parallel computers is demonstrated. Special attention is given to methods which optimize processor array use and the particular pitfalls associated with parallel implementation of the Monte Carlo technique associated with the violation of ergodicity and coupling of the system Hamiltonian dynamics with the processor update period.

1 Introduction

Oxygen ordering in the basal plane of $YBa_2Cu_3O_z$ ($6 \leq z \leq 7$) is critical to the superconducting properties of the system. The basal plane of the material can be simply described as a square basis of copper ions with sites for oxygen and structural oxygen vacancies half way between the copper ions at the (1/2,0) and (0,1/2) positions. The oxygens, quite mobile relative to the copper lattice, can take up various ordered arrangements in this plane. Of particular interest is the ordered "chain" state achieved when oxygens order in the (0,1/2) sites exclusively forming long copper-oxygen chains running along the [010] direction. This arrangement is orthorhombic by nature of the elongation imposed along the chain direction and is the superconducting phase. By contrast, a disordered distribution of oxygens over all the sites in the basal plane leads to the formation of a tetragonal, non-superconducting phase. Thus it is of key materials engineering interest to know the phase boundaries associated with the superconducting orthorhombic phase in order to fabricate this material and to characterize the dynamics of oxygen ordering to allow the creation of the best kinetic schedule for optimum production.

Experimental studies of this system are complicated by the ease of oxygen loss and the tendency of the system to be doped by many cations present as contamination or in the crucible material. In addition, at low temperatures, many complicated longer wavelength

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ordered phases are known to occur, dramatically slowing down the approach to equilibrium, and making it nearly impossible to experimentally extract reliable phase information in these regimes. This system has been shown to be well described by various statistical models of the basal plane allowing its study by computer simulation models. The type of materials science simulation technique needed to characterize the phase space of the $YBa_2Cu_3O_z$ system, as well as many other systems, operate on model systems which involve multiple data points under the influence of a single global equation of state and, hence, are ideal candidates for data parallelism. In what follows, the Monte Carlo technique is described in preparation for its parallel implementation in the study of different aspects of oxygen ordering in the $YBa_2Cu_3O_z$ superconductor. The work presented here was implemented on the MasPar 1 series (DEC mpp-12000) massively parallel SIMD architecture computer, the details of which are described elsewhere [1].

2 Monte Carlo Simulation in Materials Science

Monte Carlo simulations (see Ref. [2] for a description of the basic technique) of different systems are primarily distinguished by the selection of two factors: (1) Hamiltonian, and (2) static lattice structure and boundary conditions. In this study we will consider two different Hamiltonians proposed to describe oxygen ordering in the basal plane of $YBa_2Cu_3O_z$. Both are based on the simple Ising spin-1/2 model which employs a pair interaction energy V between particles or "spins" so described as they can take two states represented by the site occupation variable $\sigma = \pm 1$. In the case of the basal plane of $YBa_2Cu_3O_z$, a $+1$ state represents an oxygen at the site in question while a -1 state represents a vacancy. The first model is the nearest and anisotropic next-nearest neighbor Ising model proposed by Wille et al. [3] to model oxygen ordering in the basal plane of $YBa_2Cu_3O_z$

$$H = \frac{1}{2}\sum_{<NN>} V_1\sigma_i\sigma_j + \frac{1}{2}\sum_{<NNN^\dagger>} V_2\sigma_i\sigma_j + \frac{1}{2}\sum_{<NNN>} V_3\sigma_i\sigma_j \tag{2.1}$$

where the † is taken over next-nearest neighbors separated by a copper cation. Although original studies used canonical values for the interaction parameters, later work [4] succeeded in determining the V's from first-principles electronic structure calculations. Even further range interactions can be considered as in the case of the screened Coulomb potential used to explain observed oxygen superstructures in this system [5]; important to the understanding of detailed dependence of critical temperature on basal plane oxygen content, where

$$V_{ij} = V_1 \exp[-\lambda|r_i - r_j|]/|r_i - r_j| + V_2\delta_{ij}. \tag{2.2}$$

While the underlying model and lattice can remain the same, the choice of Hamiltonian dictates the approach in developing the most efficient parallel algorithm.

3 Parallel Monte Carlo - Fundamental Considerations

The central issue in constructing a correct parallel Monte Carlo algorithm is to distribute the calculation in the most efficient way amongst the processors without violating the detailed balance criterion

$$P(\Omega_i)\Pi(\Omega_i \to \Omega_j) = P(\Omega_j)\Pi(\Omega_j \to \Omega_i) \tag{3.1}$$

which ensures that simulation time averages will converge to thermodynamic ensemble averages in the limit of long times. Here, $P(\Omega_i)$ is the probability of a certain microstate Ω_i occurring and Π_{ij} is the transition probability from a state Ω_i to Ω_j. In a serial algorithm, this is achieved in a straightforward way by accepting each trial microstate in a Boltzmann distribution [2]. However, for parallel Monte Carlo algorithms, an additional constraint must be satisfied, namely, that all parallel updates are spatially independent. A set of spatially independent sites is defined as a set of sites which do not interact with each other through the action of the system Hamiltonian. For example, in the two dimensional nearest neighbor Ising model, each site interacts with its four nearest neighbors. Updating nearest neighbors in parallel leads to the creation of a chain with non-ergodic sequences in violation of the detailed balance condition (3.1). This can be understood qualitatively by considering a one dimensional chain of nearest neighbor interacting Ising spins whose ground state is ferromagnetically aligned. During an update, a site will tend to adopt the alignment of its neighbors. At some finite temperature, adjacent sites which are anti-ferromagnetically ordered will be created. If updates are executed in parallel on all sites, sites in anti-ferromagnetic sequences will oscillate between spin-up and spin-down states and never achieve an aligned ground state. This "blinking-state" or "parallel-resonance" condition has the effect of confining the system to an artificial and limited region of phase space and may lead to invalid statistics [6].

For systems which involve relatively short-range interaction, as in (2.1), the most effective method of parallel implementation which avoids parallel-resonance is *geometric decomposition* [7-9]. In this scheme, the system is divided into maximal sets of spatially independent sites which are updated in parallel. In the case of the nearest neighbor Ising model, this amounts to a "checkerboard" decomposition where every other site in each lattice direction is updated simultaneously (see, for example, Ref. [10]). Geometric decomposition has the advantage of involving a nearly direct mapping of the algorithm to the parallel array and is thus simple to implement. In the limit where interactions extend to the size of the system, each site interacts with all others and a geometric decomposition would degrade to a serial algorithm. In such cases, as in (2.2), an *algorithmic decomposition* scheme is employed [8-9]. In this approach, the parallel aspects of the algorithm are distributed to all the processors.

4 Parallel Monte Carlo - Implementation

In a standard Monte Carlo study, the system size is typically varied to elucidate the effect on the critical values. Simulating system sizes smaller than the processor array is accomplished by running on a subset of the available processors. Simulating system sizes larger than the available processors is accomplished by partitioning the larger lattice onto the smaller processor array. Partitioning is also useful in increasing the efficiency of the overall algorithm. In the checkerboard decomposition, at least half of the processors are idle at any given time. While this could constitute an acceptable use of the processor array, the number of idle processors increases rapidly with the extent of inter-site interactions such that a simulation using (2.1) with nearest and next nearest neighbor interactions would leave one fourth of the processors idle. Partitioning is accomplished by declaring sub-arrays of sites on each processor. During each step, every processor updates the equivalent site in its local partition in parallel. If the size of the partition is such that it contains the range of interactions for one site, then all processors can operate in each cycle.

The nearest and anisotropic next nearest neighbor Ising model (2.1) was implemented using a partitioning scheme. In order to avoid updating sites interacting via the Hamilto-

nian, a minimum partition size of $4x4$ was needed to contain the second nearest neighbor interaction. This parallel algorithm was used to calculate the variation of the specific heat of the system with oxygen partial pressure and temperature. The locus of the peaks in the specific heat forms the equilibrium phase diagram for the system. The high temperature transition from the ordered orthorhombic copper oxygen chain phase to the disordered tetragonal phase as well as the low temperature transitions between the two high temperature phases and an additional cell-doubled chain phase are well described [11]. It is true in general that any simulation results must be held suspect until any possibility of artifacts arising from the algorithm is eliminated, but it is doubly true in the use of parallel simulation. While ensuring that sites interacting through the Hamiltonian are not updated in parallel avoids two-step or "first-order" violations of ergodicity, under specific conditions, the coupling of the update period with the system kinetics can lead to longer period cycles or "second-order" violations of ergodicity. An example of this was found to occur at low temperatures in the simulation of oxygen ordering using the model described by (2.1). Specific heat results showed an anomalously low specific heat and an examination of the system structure revealed unusual but perfectly ordered configurations occurring consistently at an oxygen chemical potential of $-4.0k_BT/V_1$. An inspection of the step by step kinetics at the anomalous chemical potential value revealed that the system had entered into an eight step cycle leading to energy averages that were perfectly stable and, hence, lead to low specific heats. At this chemical potential, the interactions between each of the nearest neighbors perfectly balanced the contribution from chemical potential. Thus, starting from a vacant lattice, the first four updates alternately populated and left vacant the sites, and then reversed the occupation on the second four updates, leading to a stable, but artificial, eight step cycle. This phenomenon is reminiscent of the limit cycle behavior found in cellular atomata. Such an occurrence is simply avoided by introducing a degree of randomness normally supplied by thermal fluctuations at higher temperatures via a disordered initial configuration, but highlights the existence of artificial coupling between system kinetics and hardware cycles that propagate via multistep processes.

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