

A Coupled Discrete/Continuum Model for Multiscale Diffusion

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(Dated: December 8, 2004)

A method is developed to model continuum (finite element) and discrete (kinetic Monte Carlo) diffusion occurring simultaneously in connected regions of space. The two regions are coupled across an interface using an iterative domain-decomposition approach in which time-dependent boundary conditions are applied on the kMC region (concentration) and on the continuum region (flux). Evolving forward in small time increments permits iterations in the kMC region to be performed only in a narrow band near the interface. An on-the-fly convergence criterion based on the inherent fluctuations in the discrete problem is developed. Application to the decay of a Gaussian concentration profile demonstrates the accuracy and efficiency of the method. Generalizations to more-complex problems in 2d and 3d, and with spatially-varying diffusivity due to interactions or applied stress fields, are straightforward.

PACS numbers: 66.30.-h, 02.70.Bf, 02.50.Fz, 05.40.-a

Many physical processes relevant to macroscopic material behavior occur over multiple time and length scales. The need to describe detailed local phenomena and larger-scale collective phenomena simultaneously has motivated research into two broad categories of multiscale modeling. The “information passing” mode uses a detailed method, e.g. atomistics, to obtain values for parameters in a separate less-computationally-intensive framework, e.g. finite elements. The “direct coupling” mode involves simultaneous treatment of a problem using different methods in different regions as dictated by the resolution required in each region. Atomistic/continuum models, such as the quasicontinuum model [1] and various absorbing-boundary-condition models [2], couple length or time scales of deformation but with all defects resolved atomistically. The Coupled Atomistic/Discrete-Defect (CADD) [3] method includes continuum defects, such as dislocations or point defects, and the passing of the defects back and forth between atomistics and continuum. In all of these models, all defects are treated discretely; there is no transition to a continuum field description where defects disappear and are replaced by field variables such as plastic strain or concentration.

Multiscale discrete/continuum models are emerging, however. Direct-simulation Monte Carlo has been coupled to an appropriate continuum model (the Euler equation) for steady-state hydrodynamics problems [4–6]. Incompressible formulations typically employ an iterative Schwartz alternating method in which macroscopic quantities are matched in an overlapping domain. However, coupling of discrete and continuum regions over both space and time remains a challenge for non-steady-state problems. A coupling of molecular dynamics and the Navier-Stokes equation was created by O’Connell and

Thompson [7] and applied to the start-up period of a shear flow simulation. More recently, Alexander *et al* [8] developed an overlapping domain scheme to couple a discrete region characterized by the analytical solution of one-dimensional random walkers to a continuum region described by the stochastic diffusion equation solved using finite differences. Here, we describe a new discrete/continuum coupling method for non-steady flow in which a region described by the diffusion equation is connected through a surface to a region in which the diffusing entities (vacancies or impurities on a lattice) are treated explicitly by kinetic Monte Carlo (kMC), as shown schematically in Fig. 1. The present method complements that of Alexander *et al* because it can accommodate high gradients near the interface whereas the use of an overlapping domain in Alexander *et al* restricts the application to small gradients. The present method also provides a framework for extending the length and time scales of a wide range of complex problems involving explicit kinetic and energetic effects, such as the influence of imposed stresses, site-specific energetics, particle interactions, etc. for which the kMC method has been a powerful tool.

The separate treatments of continuum and discrete diffusion are well-established. Here, the continuum region is treated using the Finite Element Method (FEM) applied in a standard manner [9] to the continuum diffusion equation

$$\frac{\partial c}{\partial t} - D\nabla^2 c = 0 \quad (1)$$

with c the concentration field and D the diffusion coefficient. Eq. (1) is appropriate when the concentration varies slowly with respect to the defect hopping distance a and all diffusing entities perform independent random walks. The system of ODEs resulting from the finite element discretization of (1) is integrated using a semi-implicit time integration scheme. The kinetic Monte Carlo method is employed in the discrete region where

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the particles (vacancies or interstitial on a lattice) hop from site to site at rate Γ defined as the *total* number of hops per particle per unit time. Since the aim here is not to study complex kinetic processes but rather to connect a kMC region to a continuum region, we consider, for demonstration purposes, the diffusive hops that occur in a square lattice by particles allowed to hop to empty nearest neighbor sites with equal probability. No interactions between adjacent particles are considered and the hopping rate is independent of position so that, for low concentrations, the particles will undergo independent random walks. Thus, in a kMC simulation with N particles, time is advanced after every Monte Carlo step by $\delta t = 1/N\Gamma$. The relationship between the continuum diffusion coefficient, the kMC hopping rate Γ , and length a is well-established [10]. For a square lattice of sites,

$$D = \frac{1}{4}\Gamma a^2. \quad (2)$$

With this background, there are four main pieces to the development of our coupled method: (i) the formal domain decomposition algorithm; (ii) the implementation of boundary conditions in a kMC framework; (iii) creation of an efficient near-interface iterative approach; and (iv) establishment of an automated convergence criterion based on intrinsic fluctuations in the kMC problem and as a function of spatial resolution. Domain

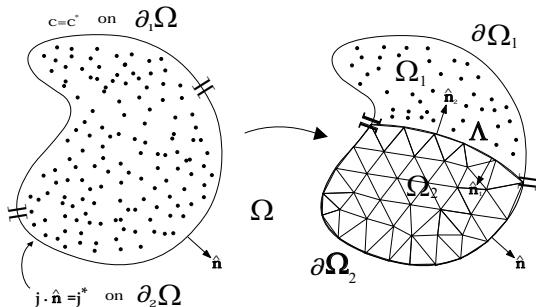


FIG. 1: Schematic of the decomposition of a discrete diffusion problem in domain Ω into a coupled problem consisting of domains Ω_1 and Ω_2 separated by an interface Λ , where Ω_1 remains discrete (e.g. kMC) while Ω_2 is approximated by the continuum diffusion equation (e.g. FEM).

decomposition methods for partial differential equations are well established [11]. We follow the spirit of this methodology but applied to a problem in which the domains are treated by different numerical methods. Consider the distribution of particles in the domain Ω shown in Fig. 1. The particles are treated explicitly and are characterized by a discrete concentration field $c(\mathbf{x}) = \sum_i \delta(\mathbf{x} - \mathbf{x}_i)$. The domain is subject to an initial concentration at $t = 0$, $c(\mathbf{x}, 0) = c_0(\mathbf{x})$ corresponding to an initial distribution of particle at positions \mathbf{x}_i^0 , and in general, concentration and flux boundary conditions on $\partial_1\Omega$ and $\partial_2\Omega$, respectively. The time evolution of the

system is governed by the rules associated with the kMC algorithm. Now consider the boundary Λ to divide Ω into sub-domains Ω_1 and Ω_2 . In Ω_1 the particles are still treated explicitly and the evolution continues to be governed by the kMC method. In Ω_2 it is assumed that the particle distribution can be adequately described by a continuum field $c(\mathbf{x})$ whose evolution is governed by the continuum diffusion equation (1). A correct solution to the entire problem is attained when the time-dependent concentration and flux on the boundary Λ are consistent between the two domains.

To achieve self-consistency along Λ , our coupled method employs an iterative scheme as follows. The k th iteration consists of running the kMC in Ω_1 with boundary condition

$$c_1^{(k)}(\mathbf{x}, t) = \lambda^{(k)}(\mathbf{x}, t), \quad \text{on } \Lambda, \text{ for } 0 \leq t \leq T \quad (3)$$

where $\lambda^{(1)}$ is a guess for the first iteration. Next, the flux through Λ , $j_{1 \rightarrow 2}^{(k)}$, is measured and applied exactly to Ω_2 by solving the diffusion equation (1) with flux boundary condition

$$-D\nabla c_2^{(k)}(\mathbf{x}, t) \cdot \hat{n} = -j_{1 \rightarrow 2}^{(k)}(\mathbf{x}, t), \quad \text{on } \Lambda \text{ for } 0 \leq t \leq T. \quad (4)$$

This generates a concentration $c_2^{(k)}|_{\Lambda}$ along Λ in Ω_2 that does not match the concentration $\lambda^{(k)}$ originally applied to Ω_1 . For iteration $k + 1$, the kMC boundary condition is taken as a weighted average of $c_2^{(k)}|_{\Lambda}$ and $\lambda^{(k)}$ characterized by a weighting factor $0 < \theta \leq 1$

$$\lambda^{(k+1)}(t) = \theta c_2^{(k)}(\mathbf{x}, t)|_{\Lambda} + (1 - \theta)\lambda^{(k)}(\mathbf{x}, t). \quad (5)$$

This iterative procedure is then repeated until convergence is obtained (see Eq. (7) below), i.e.

$$c_2^{(k)}(\mathbf{x}, t)|_{\Lambda} \approx c_1^{(k)}(\mathbf{x}, t)|_{\Lambda} \quad (6)$$

for $0 \leq t \leq T$ along the interface Λ . The solution at $t = T$ is then taken as an initial condition for a new boundary value problem for the time $T \leq t \leq 2T$, where we take $\lambda^{(1)}(\mathbf{x}, t) = c_1(\mathbf{x}, T)|_{\Lambda}$. Hence the solution can be advanced in time in *increments* of size T .

The above algorithm requires that a concentration boundary condition, Eq. (5), be applied to the kMC region, which is a non-standard procedure in most kMC implementations. We impose this boundary condition at a distinct set of times spaced by time step Δt within the time increment T . Along each boundary element composing the surface Λ and at the i th step (time $t = i\Delta t$ within the increment T) we add/subtract $n(\mathbf{x}, i\Delta t) = L_{\Lambda}(\lambda(\mathbf{x}, t) - c_1(\mathbf{x}, t)|_{\Lambda})$ particles to the boundary element, where L_{Λ} is the length of the boundary element along Λ . The flux through this element is then calculated as $j_{1 \rightarrow 2}(\mathbf{x}, i\Delta t) = n(\mathbf{x}, i\Delta t)/\Delta t L_{\Lambda}$.

In the formulation above, the kMC problem is solved iteratively over the entire kMC domain, which is undesirable and unnecessary. Convergence is obtained by matching conditions at the boundary, and particles far from the boundary cannot diffuse to the interface in time T and thus need not be considered during the iterative process. For a random walk, the probability of finding a particle a distance x from the origin at time T is a Gaussian of half-width $\bar{x} = a\sqrt{\Gamma t/2}$ [10] and so particles farther than several times \bar{x} from the interface will reach the interface in time T with very low probability. Therefore, the kMC algorithm is only iterated on those particles existing within a band of width w_b at the start of the increment, holding all other particles fixed. Once convergence at the boundary is achieved, the remaining particles outside the band region are then moved *once* for a time T . During the iterations, those particles originally within w_b may naturally move outside the band region. During the final diffusion of the particles outside the band, some of them may enter the band region. Thus, for the next time increment, there is a new set of particles located within the band. For the results presented here, $w_b = 2.2\bar{x}$.

Due to the stochastic nature of the discrete diffusion, fluctuations of the kMC concentration in the boundary elements have a magnitude $\delta c = \sqrt{N}/L_\Lambda = \sqrt{c/L_\Lambda}$ that scales with the concentration and boundary element length even in equilibrium. Thus, the residual error in concentrations between the two methods at the boundary should not be expected to be reducible far below this δc . In addition, for the highest fidelity in the discrete solution, we wish to preserve the intrinsic fluctuations in the kMC regime right up to the boundary rather than artificially forcing the boundary to a fixed concentration. We thus define convergence of the iterative scheme using a normalized error measure that accounts for the magnitude of the concentration fluctuations as

$$\varepsilon^{(k)} = \sqrt{\frac{1}{T} \int_{(i-1)T}^{iT} \frac{1}{L_\Lambda} \int_\Lambda \left(\frac{c_1^{(k)} - c_2^{(k)}}{\delta c_1^{(k)}} \right)^2 dS dt}. \quad (7)$$

With this error measure, convergence is then deemed to be achieved when $\varepsilon^{(k)} < 1$. This convergence criterion automatically accounts for the effects of both finite element size and the evolving concentration. We analyze convergence further below.

We now apply the coupled method to a demonstration problem: nominally one-dimensional diffusional flow of an initial Gaussian concentration profile. The problem consists of a continuum region extending from $x = 0$ to L_{fem} and a 2d kMC region consisting of $L_{\text{fem}} \leq x \leq L_{\text{fem}} + L_x$ and $0 \leq y \leq L_\Lambda$. The initial condition c_0 is a Gaussian distribution in x of width $\sigma = 30a$. All outer boundaries are impermeable ($j_n = 0$). Bulk diffusional flow occurs in the x direction only, so the continuum region is treated using a 1d finite element mesh and the flux into the continuum region is the total flux across L_Λ . The kMC problem includes diffusive motion perpendicular to

the bulk flow direction.

The relevant length and time are a and $1/\Gamma$, respectively; hence results are presented in the dimensionless variables and parameters $\xi = x/a$, $\tau = \Gamma t$, $\Delta\tau = \Gamma\Delta t$ and $\mathcal{T} = \Gamma T$. For the present problem, $L_{\text{fem}} = 100a$ and $L_x = 100a$ and the kMC domain has a depth of $L_\Lambda = 1,000a$. The FEM region is discretized into elements of length a ; coarsening away from the boundary is a trivial modification. The diffusion problem is computed for $0 \leq \tau \leq 18,000$. The time domain is subdivided into 300 increments of $\mathcal{T} = 60$. The kMC boundary concentration and FEM boundary flux are updated using a time step $\Delta\tau = 20$. The weighting factor is set to $\theta = 0.5$.

$\Delta\tau \geq 10$, and $0.1 < \theta < 0.9$, with $\theta = 0.5$ optimal.

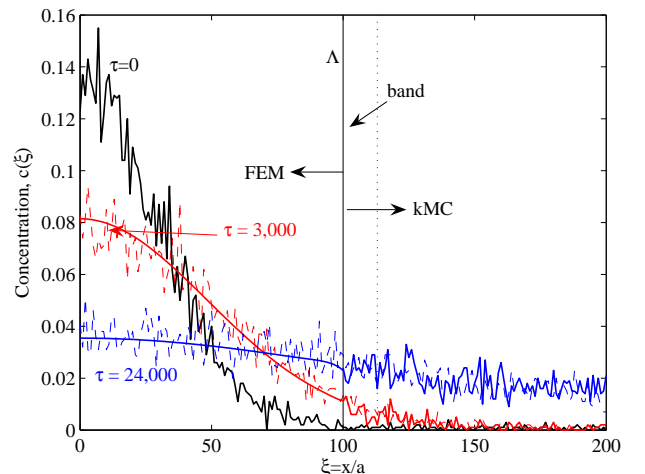


FIG. 2: Decay of an initial Gaussian concentration distribution at various dimensionless times: coupled method (solid lines); full kMC (dashed lines). A band of width $w_b = 2.2\bar{x} \approx 13a$ is shown

Fig. 2 shows the concentration profile predicted by the coupled method throughout the entire spatial domain at several points in time. Also shown for comparison are the results of a full kMC simulation of the same boundary value problem over the entire domain, a problem that requires $\sim 1 \times 10^8$ kMC steps. Agreement between the coupled and exact solutions is excellent at all times. The coupled method converges quickly, requiring an average of only 1.4 iterations per increment. This problem requires only about 25% of the computational time of the full kMC problem, a factor limited only by the comparable sizes of the two regimes. If the kMC constitutes a smaller fraction of the total problem, the gain will be even greater.

In order to examine the effects of stochastic fluctuations, system size, and our automated convergence criterion 7 on the quality of the solutions of the coupled method, we have performed similar studies for sizes $50a \leq L_\Lambda \leq 2,000a$ using a time step of $\Delta\tau = 10$. Fig. 3 shows the *normalized error* $\varepsilon^{(k)}$ vs. iteration; a size-

independent minimum is reached after just a few iterations. The inset of Fig. 3 shows the average value of the asymptotic minimum error as a function of the time step $\Delta\tau$. The error can be reduced below $\varepsilon = 1$ for larger $\Delta\tau$ values because such larger values smooth out the measured flux from the kMC. Thus a more stringent residual error may be demanded when $\Delta\tau > 10$ but, as noted above, such solutions exhibit unnaturally low fluctuations at the boundary and do not actually improve the overall solution in the two domains. For $\Delta\tau < 10$, an asymptotic error $\varepsilon > 1$ is achieved that we believe is due to sampling of the kMC boundary concentration at a rate that is just too fast (less than 10 hops per particle) for reasonable correspondence between the discrete and continuum problems. Hence, by setting convergence at $\varepsilon = 1$ the algorithm accepts solutions that exhibit the natural fluctuations associated with the system size and converges quickly for all system sizes L_Λ , time steps $\Delta\tau \geq 10$, time increments \mathcal{T} , and weighting factors θ .

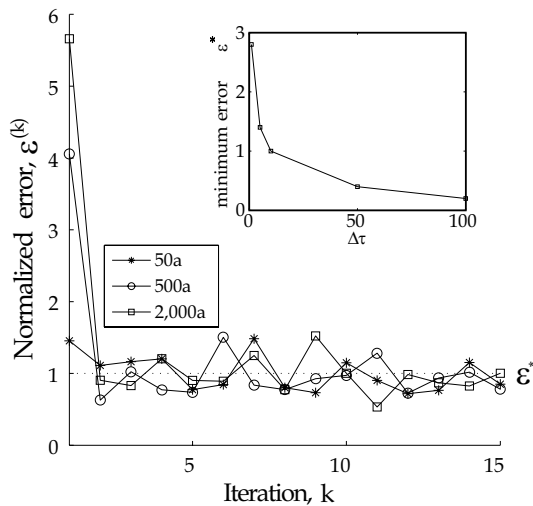


FIG. 3: Normalized error as a function of iteration number k , for system sizes $50a \leq L_\Lambda \leq 2,000a$, and time step $\Delta\tau = 10$. The normalized error quickly reaches a size-independent asymptotic minimum value that varies with time step $\Delta\tau$ as shown in the inset.

There are three main reasons for choosing the current scheme wherein the kMC region has an imposed concentration boundary condition as opposed to one in which concentration is measured from the kMC and applied to the FEM: (i) flux continuity is satisfied by construction and leads directly to mass conservation, a much more important condition than continuity of concentration; (ii) by measuring flux and not concentration on the kMC boundary, we avoid the added computation of averaging over an area near the boundary, and (iii) the processes of applying a concentration and measuring the flux are incorporated in a simple subroutine.

Here, the basic algorithm for coupling kMC and FEM regions has been laid out and tested in one dimension. Extensions to two dimensions are straightforward since the kMC is already two-dimensional and we have demonstrated convergence for element sizes down to $L_\Lambda = 50$. Extensions to three dimensions are straightforward, and convergence is expected for element areas down to $A_\Lambda = 50$, i.e. element edge lengths of $L_\Lambda = 5 - 10$ that approach the fundamental length scale of the discrete hopping. Furthermore, extensions to incorporate near-neighbor interactions, applied fields, etc. can be easily implemented in the coupled formulation whenever an appropriate continuum diffusion equation is available with, for instance, a concentration- or field-dependent diffusion coefficient or driving force (chemical potential). Use of the coupled scheme here in the presence of such effects can significantly increase the flexibility of simulations by allowing for the imposition of remote boundary conditions in non-periodic systems and by localizing the highly-intensive computations associated with the kMC exclusively to regions that are not describable by the continuum approximation (e.g. high concentrations, near neighbor interactions, adatom-step interactions, high local fields, etc.).

Acknowledgments

The authors gratefully acknowledge support of this work by the US AFOSR through Grant F49620-99-1-0272 through the MURI program Virtual Design and Testing of Materials: A Multiscale Approach.

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