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Coarse-grained molecular dynamics and the atomic limit of finite elements

Robert E. Rudd* and Jeremy Q. Broughton†
Complex Systems Theory Branch, Naval Research Laboratory, Washington, D.C. 20375-5345

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We develop a technique for simulation of the mechanics of micron-scale solid systems: coarse-grained molecular dynamics (CGMD). It captures the important atomistic effects without the computational cost of conventional molecular dynamics (MD). The CGMD equations of motion are derived directly from finite temperature MD through a statistical coarse graining procedure, so they agree with MD as the mesh size is reduced to the atomic scale. This allows a seamless coupling of length scales. The use of efficient CGMD in peripheral regions extends atomistic simulation to much larger systems than amenable to MD alone. [S0163-1829(98)52134-0]

The physical world is replete with phenomena on different scales that are only weakly coupled. Physics would be totally intractable were this not so. There are many systems, however, which depend inherently on physics on multiple scales. These pose notoriously difficult theoretical problems. Turbulence, crack propagation, and critical phenomena at nontrivial fixed points are prime examples. In this paper we focus on another multiscale problem, mesoscopic elastic dynamics, and we formulate an effective theory which couples micron-scale boundary conditions to physics at the atomic scale.

The mesoscopic elastic problem is this: there are interesting solid-state systems whose elastic properties can be modeled neither by finite elements because they are too small, nor by atomistics because they are too large. Examples include technologically important micro-electro-mechanical systems (MEMS) being constructed now, and mesoscopic systems of crack propagation and delamination. To be more precise, dynamical regions smaller than 1 μm are affected by atomic scale physics which causes departures from continuum elastic theory,[1] and dynamical regions larger than 0.1 $\mu\mathrm{m}^3$ exceed the current limit of about one hundred million atoms for atomistic simulation of silicon on a supercomputer.[2] The challenge is to develop a robust simulation technique which captures the important atomistic effects without the computational cost of a brute force atomistic simulation for the entire system.

Consider the standard finite element (FE) approach to this problem.[3] A mesh of varying coarseness is chosen, adaptively or by fiat, such that no one region contributes disproportionately to the error. Typically errors result from large strain gradients violating the discrete expression for the integral of the elastic energy density of a continuous medium. Mesh refinement improves this approximation. However, as the mesh size approaches atomic dimensions, the constitutive equations have significant errors, and further refinement alone does not help. One proposal to improve this situation replaces the FE equations of motion on regions of the mesh that are atomic-sized with MD equations of motion and implements a consistent hand shaking between the MD and FE regions.[4,5] Another hybrid FE/MD proposal is the quasicontinuum technique, a zero temperature relaxation technique.[6]

In this paper, we present a method of constructing scale-dependent constitutive equations suitable for a mesh which is atomic sized in some regions. In these regions it is guaranteed to reproduce the atomistic equations of motion. This enables MD regions to be coupled seamlessly to regions of generalized FE, bringing the full power of MD to bear on important parts of the system without the computational overhead of MD in other large, but physically less complex regions. The procedure, coarse grained molecular dynamics (CGMD), is based on a statistical coarse graining prescription.

Suppose we are given a microscopic potential energy expression describing the motion of atoms in a solid, crystalline or amorphous, and a coarse grained (CG) mesh partitioning the solid into cells. The mesh size may vary, so that in important regions a mesh node is assigned to each equilibrium atomic position, whereas in other regions the cells contain many atoms and the nodes need not coincide with atomic sites. We propose a method to produce equations of motion for a mean displacement field defined at the nodes. In particular, we define the conserved energy functional for the CG system as a constrained ensemble average of the atomistic energy under fixed thermodynamic conditions. The equations of motion are Hamilton's equations for this energy functional.

The classical ensemble must obey the constraint that the position and momenta of the atoms are consistent with the mean displacement and momentum fields. Let the displacement of atom μ be $\mathbf{u}_\mu = \mathbf{x}_\mu - \mathbf{x}_{\mu 0}$ where $\mathbf{x}_{\mu 0}$ is its equilibrium position. The displacement of mesh node j is an average of the atomic displacements

$$\mathbf{u}_j = \sum_\mu f_{j\mu} \mathbf{u}_\mu , \tag{1}$$

where $f_{j\mu}$ is a weighting function, related to the microscopic analog of FE interpolating functions below. An analogous relation is implied for the momenta $\mathbf{p}_\mu$. Since the nodal displacements are fewer or equal to the atomic positions in number, fixing the nodal displacements and momenta does not (necessarily) determine the atomic coordinates entirely. Some subspace of phase space remains, corresponding to degrees of freedom that are missing from the mesh. We define the CG energy as the average energy of the canonical ensemble on this constrained phase space:

$$E(\mathbf{u}_k,\dot{\mathbf{u}}_k)=\langle H_{MD}\rangle_{\mathbf{u}_k,\dot{\mathbf{u}}_k} \tag{2}$$

$$=\int d\mathbf{x}_\mu d\mathbf{p}_\mu H_{MD} e^{-\beta H_{MD}}\Delta/Z, \tag{3}$$

$$\Delta=\prod_j \delta\left(\mathbf{u}_j-\sum_\mu \mathbf{u}_\mu f_{j\mu}\right) \quad \delta\left(\dot{\mathbf{u}}_j-\sum_\mu \frac{\mathbf{p}_\mu f_{j\mu}}{m_\mu}\right), \tag{4}$$

where $\beta=1/(kT)$ is the inverse temperature, Z is the partition function and $\delta(\mathbf{u})$ is a three-dimensional delta function. The delta functions enforce the mean field constraint (1). Note that Latin indices, $j,k,\ldots,$ denote mesh nodes and Greek indices, $\mu,\nu,\ldots,$ denote atoms. The energy (3) is computed below [Eq. (8)].

When the mesh nodes and the atomic sites are identical, the CGMD equations of motion agree with the atomistic equations of motion. As the mesh size increases some short-wavelength degrees of freedom are not supported by the coarse mesh. These degrees of freedom are not neglected entirely, because their thermodynamic average effect has been retained. This approximation is expected to be good provided the system is initially in thermal equilibrium, and changes to the system would only produce adiabatic changes in the missing degrees of freedom. As long as this condition is satisfied, the long wavelength modes may be driven out of equilibrium without problems.

The CG energy (4) may be computed using standard techniques. We have derived the expression in closed form for a monatomic harmonic solid. We take the form of the atomistic Hamiltonian to be

$$H_{MD}=\sum_\mu \frac{\mathbf{p}_\mu^2}{2m}+\sum_{\mu,\nu}\frac{1}{2}\mathbf{u}_\mu\cdot D_{\mu\nu}\cdot\mathbf{u}_\nu, \tag{5}$$

where $D_{\mu\nu}$ is the dynamical matrix. It acts as a tensor on the components of the displacement vector at each site. We define the mass matrix by the matrix inverse

$$M_{jk}=m\left(\sum_\mu f_{j\mu}f_{k\mu}\right)^{-1} \tag{6}$$

and the stiffness matrix by the matrix inverse

$$K_{jk}=\left(\sum_{\mu\nu} f_{j\mu}D^{-1}_{\mu\nu}f_{k\nu}\right)^{-1}. \tag{7}$$

The CG energy (3) for a monatomic harmonic solid of N atoms coarse grained to N_{node} nodes is computed to be

$$E(\mathbf{u}_k,\dot{\mathbf{u}}_k)=U_{int}+\frac{1}{2}\sum_{j,k}(M_{jk}\dot{\mathbf{u}}_j\cdot\dot{\mathbf{u}}_k+\mathbf{u}_j\cdot K_{jk}\cdot\mathbf{u}_k), \tag{8}$$

where $U_{int}=3(N-N_{node})kT$. The energy contains terms representing the average kinetic and potential energies, plus the thermal energy term expected from the equipartition theorem for the modes that have been integrated out. This Hamiltonian is easily generalized to polyatomic solids, where the optical modes may be coarse grained in several ways to represent different physics. Also, we have formulated CGMD for an underlying anharmonic Hamiltonian in perturbation theory, assuming negligible diffusion in the CG region. Here the higher modes do not decouple, and energy may flow to and from the internal ensemble, contributing to thermal expansion, etc. Those results will be presented elsewhere.[7]

The stiffness matrix K_{ij} is to be computed once at the start of a simulation, and it remains unaltered during the subsequent dynamics. It does not matter whether atoms vibrate across cell boundaries, as long as diffusion is negligible. An efficient computation of K_{ij} is achieved through a normal mode decomposition of $D_{\mu\nu}$. Use of Bloch symmetry reduces the size of the dynamical matrix to be inverted to the size of the supercell of which the system is comprised. This allows the simulation of billion atom crystals on desktop workstations without approximation beyond those presented here. Note that for $T\neq 0$ the finite temperature dynamical matrix should be used for $D_{\mu\nu}$. This ensures a consistent thermodynamics. For example, in ergodic systems the time average of the kinetic energy term in the CG energy (8) is related to the temperature through a virial expression. In general, the dynamical matrix may depend on other macroscopic parameters, as well, such as slowly varying external magnetic and electric fields. $D_{\mu\nu}$ should be evaluated under these conditions. Also note that while the harmonic approximation may be good in peripheral regions, it may not be appropriate for the important regions. We have shown that the CGMD and MD equations of motion agree in regions where the mesh coincides with the atomic sites. In these regions, the full MD potential may, and should, be restored, so that effects such as diffusion and dislocation are allowed.

Several comments about the CG energy are in order. First, the mass and stiffness matrix definitions involve matrix inverses. This is somewhat ill defined for the stiffness matrix because $D_{\mu\nu}$ is singular, due to the zero modes (gapless acoustic phonons). Since there are two inverses in Eq. (7), the matrix K_{ij} is finite after a suitable regularization. For instance,

$$K_{jk}=\lim_{\epsilon\to 0}\left(\sum_{\mu\nu} f_{j\mu}(D_{\mu\nu}+\epsilon I_{\mu\nu})^{-1}f_{k\nu}\right)^{-1}, \tag{9}$$

where $I_{\mu\nu}$ is the identity matrix. We have developed a more efficient and numerically stable regularization which will be presented elsewhere.[7]

The zero modes are not integrated out, so a short-ranged $D_{\mu\nu}$ results in a short-ranged K_{ij}. On the other hand, a nearest-neighbor $D_{\mu\nu}$ does not generally produce a nearest-neighbor K_{ij}, except where the mesh is atomic-sized. The stiffness matrix elements typically decrease exponentially with separation, so the effective interaction is short ranged but not nearest neighbor. This is an important point, since it is this quality that improves the CGMD phonon spectrum.

In addition to the general framework we have presented for CGMD, a specific choice of the weighting functions is required for calculations. In general, suppose we have a set of interpolating functions $\{N_j(\mathbf{x})\}_{j=1}^{N_{node}}$ that are linearly independent at the mesh nodes [$\det N_j(\mathbf{x}_k)\neq 0$ where $\mathbf{x}_k$ is the location of the kth node]. Then we can define the displacement field $\mathbf{u}(\mathbf{x})=\Sigma\mathbf{u}_jN_j(\mathbf{x})$. Also, given any list of atomic

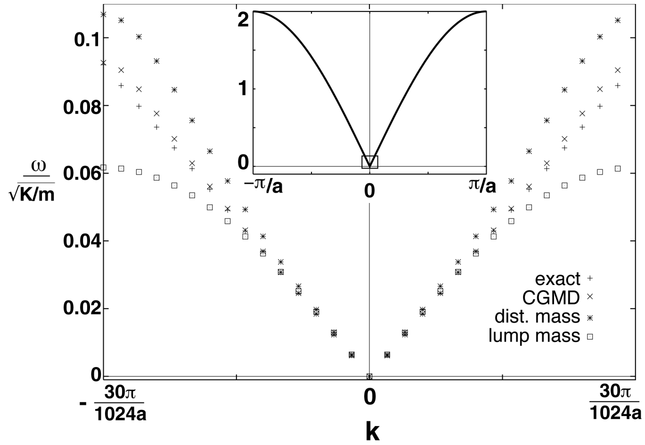

FIG. 1. The phonon spectra shown result from various treatments of a ring of 1024 atoms and an incommensurate regular mesh of 30 nodes. The inset is the exact phonon spectrum, where the coarse-grained spectrum occupies the small box near the zone center. The error in the CGMD spectrum is much less than that of the FE spectra.

displacements we can find the displacement field represented on the CG mesh which best fits these data in the least-squares sense:

$$\chi^2 = \sum_\mu \left| \mathbf{u}_\mu - \sum_j \mathbf{u}_j N_{j\mu} \right|^2, \tag{10}$$

where $N_{j\mu} = N_j(\mathbf{x}_{\mu 0})$. This χ^2 error is minimized by $\mathbf{u}_j = \Sigma_\mu f_{j\mu} \mathbf{u}_\mu$ [cf. (1)], where

$$f_{j\mu} = \sum_k \left(\sum_\nu N_{j\nu} N_{k\nu} \right)^{-1} N_{k\mu}. \tag{11}$$

This defines the weighting function $f_{j\mu}$ of (1) in terms of the interpolating function $N_j(\mathbf{x})$.

The continuum limit of the elastic energy is proportional to an integral of the square of the strain, $\boldsymbol{\nabla}\mathbf{u}$. To avoid pathologies in this limit, the displacement field $\mathbf{u}(\mathbf{x})$ should be continuous, but not necessarily smooth. The usual linear interpolation functions of finite elements are the simplest such choice. They are defined such that $N_j(\mathbf{x})$ is 1 at node $\mathbf{x}_j$, it goes linearly to zero at the nearest-neighbor nodes, and it vanishes outside of the nearest cells. These functions have the desirable properties of locality and ease of use.

Other choices of interpolating functions are possible, such as higher order polynomials. One basis we have found useful is the set of the longest wavelength normal modes. It provides a check of the CG Hamiltonian (3), since it is the optimal choice for a regular CG mesh—the phonon spectrum comes out exactly correct, apart from the missing short-wavelength modes.

As a proof of concept, we consider one-dimensional chains of atoms with periodic boundary conditions. The first test is the phonon spectrum for atoms with harmonic interactions coarse grained to a regular, but not necessarily commensurate mesh. The normal modes are plane waves both on the underlying ring of atoms and on the CG mesh. The wave vector $\mathbf{k}$ is a good quantum number for both. The nonzero terms of the dynamical matrix are of the form: $D_{\mu\mu} = 2K$, $D_{\mu,\mu\pm 1} = -K$. Figure 1 shows the resulting phonon spectra in four cases: exact, CGMD, distributed mass FE and lumped mass FE.[8] The latter two use the long wavelength elastic constants. The spectra are for a periodic chain of 1024 atoms with lattice constant a coarse grained to 30 nodes.

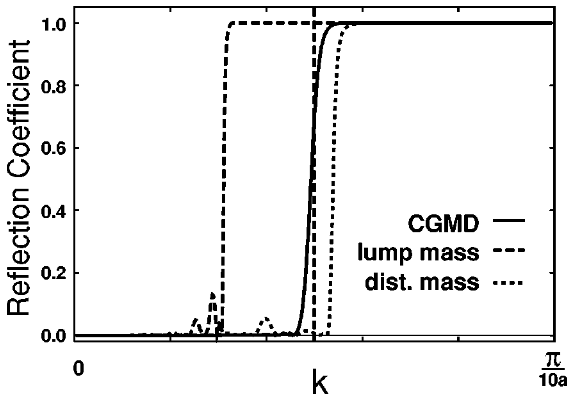

FIG. 2. A comparison of the reflection of elastic waves from a CG region in three cases: CGMD and two varieties of FE. The dashed line marks the natural cutoff (see text).

Figure 1 shows that CGMD gives a better approximation to the true phonon spectrum than the two kinds of FE do. All three do a good job at the longest wavelengths, as expected, but CGMD offers a higher order of accuracy. The relative error for CGMD is $\mathcal{O}(k^4)$ while that of the two versions of FE is only $\mathcal{O}(k^2)$. At shorter wavelengths, there are significant deviations from the exact spectrum. The worst relative error of CGMD is about 6%, better by more than a factor of 3 than that for FE. This improvement is made possible by the longer-ranged interactions of CGMD as compared to FE. The continuity condition satisfied by linear interpolation is enough to ensure that the hydrodynamic modes $(k \sim 0)$ are well modeled, but the lack of continuity of the derivatives shows up as error in the spectrum of the modes away from the zone center. This error vanishes for the smooth, nonlocal basis consisting of the longest wavelength normal modes. It turns out that the CGMD error at the CG zone boundary is relatively small (less than 1%) for technical reasons. Also note that even though the number of atoms varies from cell to cell in the incommensurate mesh, the CGMD spectrum is free of anomalies. Other computations have shown that CGMD with linear interpolation is well behaved on irregular meshes, as well.

We have computed an analytic expression for the CGMD spectrum on a commensurate mesh:

$$\omega(k) = 2\sqrt{\frac{K}{m}} \left(\frac{\Sigma_{p=0}^{n'-1} \sin^{-4}\left(\frac{1}{2}ka + \frac{\pi p N_{node}}{N} \right)}{\Sigma_{p=0}^{n'-1} \sin^{-6}\left(\frac{1}{2}ka + \frac{\pi p N_{node}}{N} \right)} \right)^{1/2}, \tag{12}$$

where $n' = N/N_{node}$. This formula shows the contribution of many modes of the underlying crystal to each CGMD mode, resulting from the choice of interpolation functions which have many normal mode components. Near the center of the CG Brillouin zone, a single mode $(p=0)$ dominates the sums (12), whereas near the boundary $[k \approx N_{node}\pi/(Na)]$, many modes contribute to keep the CGMD spectrum close to the true spectrum which is not smooth at the boundary.

Most of the applications that we envision for CGMD are dynamical and have varying mesh size. For example, in studies of crack propagation the far-field regions away from the crack may be coarse grained. For these applications it is important that elastic waves generated at the crack tip are able to propagate into the CG region. One source of finite-size effects are waves which are reflected back unphysically from boundaries or artificial interfaces.[9] This also produces a nonzero Kapitza resistance, which may cause uneven heating across the interface. Of course, a stationary system started in thermal equilibrium remains at a constant, uniform temperature given a reasonable measure of temperature in the CG region.

The natural measure of the ability of waves to propagate from an atomistic region into a CG region is the S matrix of scattering theory, or in one dimension, the transmission and reflection coefficients, $\mathcal{T}$ and $\mathcal{R}$, respectively. In Fig. 2 we plot $\mathcal{R}(k)$ for scattering from a CG region of 72 nodes representing 652 atoms in the middle of an infinite harmonic chain of atoms. The cell size increases smoothly in the CG region, as it should, to a maximum of $N_{\max}=20$ atoms per cell. In all three cases shown $\mathcal{R}$ vanishes in the long wavelength limit, and it goes to unity as the wavelength becomes smaller than the mesh spacing—a coarse mesh cannot support short wavelength modes.

At intermediate wavelengths, however, the behavior is quite different. The two finite element simulations show anomalous phonon resonances, corresponding to unphysical excitations in the CG region. These are almost completely absent from CGMD. Also, whereas the cutoff for CGMD occurs at $k=\pi/(N_{\max}a)$, exactly where it should be, the two finite element cutoffs are shifted. CGMD also reduces unwanted scattering of waves traveling from the CG region into the atomistic region, as compared to FE, but we have not shown the plot for lack of space. Of course, any long-lived short-wavelength elastic waves that would reflect from the CG region must be handled as well. We are developing a mechanism based on resonant Nosé-Hoover chains[10] to absorb and thermalize these modes.[7] For it to work, the underlying simulation must be well behaved and predictable, and this is exactly what CGMD offers.

CGMD is a technique for simulation of the mechanics of micron-scale systems. CGMD captures the important atomistic effects without the computational cost of an ordinary atomistic simulation. This is made possible by a seamless coupling of length scales: important regions of the system may be modeled with MD, while peripheral regions are coarse grained for efficiency. The CGMD equations of motion smoothly match those of MD as the mesh size is reduced to the atomic scale. Much larger systems may be modeled with CGMD than would otherwise be possible in an atomistic simulation.

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*Also at SFA, Inc., 1401 McCormick Drive, Largo, MD 20774. Electronic address: rudd@dave.nrl.navy.mil

†Electronic address: broughto@dave.nrl.navy.mil

[1] J. Q. Broughton, C. A. Meli, P. Vashishta, and R. K. Kalia, Phys. Rev. B **56**, 611 (1997).

[2] F. F. Abraham and J. Q. Broughton, Comput. Mater. Sci. **10**, 1 (1998).

[3] See, for example, O. C. Zienkiewicz and R. L. Taylor, *The Finite Element Method*, 4th ed. (McGraw-Hill, New York, 1991), Vol. II.

[4] S. Kohlhoff, P. Gumbsch, and H. F. Fischmeister, Philos. Mag. A **64**, 851 (1991).

[5] F. F. Abraham, J. Q. Broughton, E. Kaxiras, and N. Bernstein, Comput. Phys. (to be published).

[6] E. B. Tadmor, M. Ortiz, and R. Phillips, Philos. Mag. A **73**, 1529 (1996).

[7] R. E. Rudd and J. Q. Broughton (unpublished).

[8] The FE lumped mass matrix is a diagonal approximation of the true distributed mass matrix (cf. Ref. 3).

[9] F. F. Abraham, D. Brodbeck, R. A. Rafey, and W. E. Rudge, Phys. Rev. Lett. **73**, 272 (1994).

[10] G. J. Martyna, M. L. Klein, and M. Tuckerman, J. Chem. Phys. **97**, 2635 (1992).