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Variational Boundary Conditions for Molecular Dynamics Simulations of Solids at Low Temperature

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Abstract. Boundary conditions for molecular dynamics simulation of crystalline solids are considered with the objective of eliminating the reflection of phonons. A variational formalism is presented to construct boundary conditions that minimize total phonon reflection. Local boundary conditions that involve a few neighbors of the boundary atoms and limited number of time steps are found using the variational formalism. Their effects are studied and compared with other boundary conditions such as truncated exact boundary conditions or by appending border atoms where artificial damping forces are applied. In general it is found that, with the same cost or complexity, the variational boundary conditions perform much better than the truncated exact boundary conditions or by appending border atoms with empirical damping profiles. Practical issues of implementation are discussed for real crystals. Application to brittle fracture dynamics is illustrated.

Key words: Boundary condition; molecular dynamics; phonon reflection; crack propagation.

1 Introduction

The main purpose of this paper is to present a systematic study of the boundary conditions for molecular dynamics (MD) simulation of crystalline solids at low temperature. We have in mind two kinds of problems: either the MD is done in isolation with some experimental loading conditions applied to the boundary of the MD domain, or it is coupled with a continuum model outside the MD region. It has been realized that in both cases, at low temperature, the key issue is to eliminate the reflection of phonons at the boundary of the MD region or the continuum-MD

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interface. Here we will present both the theoretical framework and the numerical procedure for constructing boundary conditions that accomplish this.

The problem, as formulated above, closely resembles the problem of absorbing boundary conditions for the numerical computation of wave propagation at the far field boundary where the computational domain is truncated [14,15]. Indeed our work is very much inspired by the work in that area. In the physical world, waves know of no boundaries as they propagate. But to compute them numerically, one has to truncate the domain somewhere in order to have a computational domain of finite size. At this artificially created boundary, some numerical boundary conditions have to be used for any kind of numerical algorithms. Ideally the numerical boundary conditions should be such that the computed solutions closely approximate the physical solutions in the infinite medium. Since there are no sources of waves outside the computational domain, this amounts to requiring that the waves are not reflected at the artificial boundary.

In principle, one can write down exact reflectionless boundary conditions. In section 2, we include a concise derivation of such exact boundary conditions. In practice, however, these exact boundary conditions are of little use since they are *nonlocal both in space and in time*. Furthermore, the influence kernels (also known as response functions, influence matrices, time history kernels etc) in these boundary conditions decay rather slowly. Therefore since the 70's, much effort has gone into finding approximate but *local* boundary conditions that eliminate wave reflection to high order accuracy. Most well-known among such boundary conditions are the absorbing boundary conditions proposed by Engquist and Majda [14, 15], which were based on approximating the Fourier symbols associated with the exact boundary conditions, by either Taylor expansion or Padé approximation in the regime of near normal incidence.

Our problem for MD is very similar in spirit. The main issue is again the formulation of boundary conditions so that phonons, the discrete lattice waves, are not reflected at the boundary. Indeed one can also write down exact boundary conditions that accomplish this. This was first done by Adelman and Doll for the simple one-dimensional discrete wave equation [1]. W. Cai et al. discussed how one can in principle obtain such exact boundary conditions in the general case via numerical computations. This amounts to computing the response functions for the boundary [6]. Karpov, Liu, Park, Wagner and co-workers continued with the path suggested by Adelman and Doll, and extended that formalism to general crystal structures [22, 27, 32].

Even though one can in principle obtain such exact boundary conditions, they have the same difficulty as for the wave equation: they are nonlocal in space and time, and the influence kernels decay rather slowly. Experience with the wave equations suggests that these exact boundary conditions may not be the most efficient tools for numerical computations. This is particularly an issue in coupled continuum-MD simulations where we expect the MD region to change as the computation proceeds. Therefore as for wave equations, it is of considerable practical interest to find approximate boundary conditions which are local. This idea was first pursued in the work of E and Huang [11, 12] for simplified models, and the present paper continues on that path.

At this point, it is important to note a crucial difference between the wave equation and MD: the wave equation is continuous and for that reason, as long as the waves are fully resolved by the numerical grid, we can use small wavenumber approximations. In contrast, MD is discrete and the phonon spectrum spreads over all wavenumbers. The boundary conditions that we devise