Boundary conditions for molecular dynamics

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joint work with
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Motivation

Holian and Ravelo PRB 1994

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Boundary conditions for molecular dynamics
Outline

- Exact boundary conditions
- Phonon reflection
- Variational formulation
- Examples
- Application to fracture simulation
Problem setup

\[ m\ddot{u}_i = -\sum_j D_{ij}u_j \]
\[ m\ddot{u}_i = -\frac{\partial V}{\partial x_i}. \]
MD boundary condition: a 1D example

Exact boundary condition can be obtained by solving the **half space** problem:

\[
\begin{align*}
\ddot{u}_j &= u_{j+1} - 2u_j + u_{j-1}, \quad j \leq 0 \\
u_j(0) &= 0, \quad v_j(0) = 0, \quad j \leq 0 \\
u_b(t) &= u_1(t).
\end{align*}
\]
1D example: exact boundary condition

Exact boundary condition (Adelman et. al. 1974, 1976):

\[ u_0(t) = \int_0^t \beta(t - s)u_1(s)ds, j \leq 0 \]

\[ \beta(t) = \frac{J_2(2t)}{t}. \]
Faster decay by using more atoms

\[ u_0 = \sum_{j=1}^{J} \int_0^t \alpha_j(t-s)u_j(s)ds \]

\[ \alpha(t) \sim \frac{C}{t^J}, \quad t \to +\infty. \]
General lattice

Linearized equation of motion,

\[ M \ddot{u}_{i,j,k} = \sum_{l,m,n} D_{i-l,j-m,k-n} u_{l,m,n}, \]

Fourier transform in the \( j \) and \( k \) direction,

\[ M \ddot{U}_i(\eta, \zeta) = \sum_l D_{i-l}(\eta, \zeta) U_l(\eta, \zeta). \]

\[ U = F_{j \rightarrow \eta, k \rightarrow \zeta}[u], \quad D = F_{j \rightarrow \eta, k \rightarrow \zeta}[D]. \]

**Exact boundary condition:**

\[ u_{0,j,k}(t) = \sum_m \sum_n \int_0^t \theta_{l,j-m,k-n}(t - \tau) u_{l,m,n}(\tau) d\tau. \]
Existing work

- W. Cai, M. de Koning, V. V. Bulatov and S. Yip 2000,
- G.J. Wagner, G.K. Eduard and W.K. Liu 2004,
Practical issue

**Exact boundary conditions:**
- **nonlocal** in both space and time
- premature truncation leads to large reflection
- not feasible in a multiscale method

**Objectives:**
- local boundary conditions
- given stencil, find the BC with minimal phonon reflection
- take into account of external loading

An analogy: boundary condition for the wave equation: ABC (Engquist and Majda 1979), and many other methods.
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Phonon spectrum

Harmonic approximation:

\[ m_i \ddot{u}_i = - \sum_j D_{i-j} u_j. \]

Dynamic matrix:

\[ M \mathcal{D}(k) = \sum_j D_j e^{-i r_j \cdot k}, \]

Phonon spectrum

\[ \mathcal{D}(k) \varepsilon_s(k) = \lambda_s \varepsilon_s(k). \]
Brillouin zone: triangular lattice

the first Brillouin zone

Phonon spectrum

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Brillouin zone

Symmetry of the spectrum:
\[ \omega(k) = \omega(Pk), \epsilon(Pk) = P \epsilon(k). \]

Integration over the BZ: K point method (Monkhorst and Pack 1976).

The symmetry of the grid points substantially reduces the computation.
Brillouin zone

Symmetry of the spectrum:

\[ \omega(k) = \omega(Pk), \quad \varepsilon(Pk) = P\varepsilon(k). \]
Brillouin zone

Integration over the BZ: K point method (Monkhorst and Pack 1976).

\[
\begin{align*}
    u_i & = \frac{(2i - nq - 1)}{(2nq)} \\
    k_{ijl} & = u_i b_i + u_j b_j + u_l b_l.
\end{align*}
\]

The symmetry of the grid points substantially reduces the computation.
Phonon reflection

Taborek and Goodstein 1979

Incident and reflected waves
Phonon reflection:

\[ k' - (k' \cdot n) n = k^R - (k^R \cdot n) n \]

\[ \omega_s(k') = \omega_s'(k^R). \]

Let \( \lambda = e^{i(k^R \cdot n) a_n} \)

\[ \det(D(k^R) - \omega(k') I) = 0, \]

leads to a polynomial for \( \lambda \). The roots of the polynomial come in pairs \((\lambda, 1/\lambda^*)\). The degree of the polynomial: \( Nd \times Ne \times Na \).

The wavenumber:

\[ k^R_{s,s'} \cdot n = k_r + ik_i. \]
Phonon reflection: \textit{reflection coefficients}

\textbf{Boundary condition:}

\[ u_0(t) = \sum_j \int_0^{t_0} \alpha_j(\tau)u_j(t - \tau) d\tau. \]
Phonon reflection: *reflection coefficients*

**Boundary condition:**

\[ u_0(t) = \sum_j \int_0^{t_0} \alpha_j(\tau)u_j(t - \tau)d\tau. \]

**Incident and reflected waves:**

\[ u_j(t) = c_se^{i(r_j \cdot k - \omega_s t)}\varepsilon_s(k) \]
Phonon reflection: *reflection coefficients*

**Boundary condition:**

\[
u_0(t) = \sum_j \int_0^t \alpha_j(\tau)u_j(t - \tau) d\tau.
\]

**Incident and reflected waves:**

\[
u_j(t) = c_s e^{i(r_j \cdot \mathbf{k} - \omega_s t)} \varepsilon_s(\mathbf{k}) + c_{ss'}^R e^{i(r_j \cdot \mathbf{k}_{ss'}^R - \omega_{s'} t)} \varepsilon_{s'}(\mathbf{k}_{ss'}^R)
\]
Phonon reflection: *reflection coefficients*

**Boundary condition:**

\[
\mathbf{u}_0(t) = \sum_j \int_0^{t_0} \alpha_j(\tau) \mathbf{u}_j(t - \tau) d\tau.
\]

**Incident and reflected waves:**

\[
\mathbf{u}_j(t) = c^I S e^{i(\mathbf{r}_j \cdot \mathbf{k} - \omega_s t)} \varepsilon_s(\mathbf{k})
\]

\[
+ c^R S e^{i(\mathbf{r}_j \cdot \mathbf{k}^R - \omega_s' t)} \varepsilon_{s'}(\mathbf{k}^R)
\]

\[
c^R = R c^I,
\]

**Linear system:**

\[
(I - \mathcal{A}(\mathbf{k}) ) \varepsilon_s(\mathbf{k}) + \sum_{s'} R_{ss'}(I - \mathcal{A}(\mathbf{k}_{ss'}^R)) \varepsilon_{s'}(\mathbf{k}).
\]
Thermal flux

Energy flux at the atomic scale:

\[ J = \frac{1}{2} \sum (\dot{u}_i + \dot{u}_j) D_{i-j}(u_i - u_j) r_{ij}. \]

Convert to Fourier space:

\[ J = J^I + J^R. \]

\[ J^R = \int |c_{ss'}^R|^2 \omega_{s'} \nabla \lambda_{s'}(k) dk, \]

\[ J_n^R = 2 \sum_s \int_{k \in BZ, k \cdot n \leq 0} |\sum_{s'} c_{s'}^I R_{ss'}|^2 \omega_s^2 (\nabla \omega_s \cdot n) dk. \]

This is the thermal flux due to the applied boundary condition.
Variational boundary conditions

Discrete boundary condition:

$$u_0^{n+1} = \sum_{j=1}^{J} \sum_{m=1}^{M} \alpha_j^m u_j^{n-m} \Delta t,$$

Variational formulation:

$$\min_{\{\alpha_j^m\}} \sum_s \int_{k \in BZ, \, k \cdot n \leq 0} \sum_{s'} |R_{ss'}|^2 |(\nabla \omega_s \cdot n)| \, dk$$

subject to certain constraints.
Symmetry properties

\[ \tilde{\alpha}_j' = P \alpha_j P^T, \]

Reflection matrices:

\[ R(k; \{\alpha_j\}) = R(P^T k; \{\alpha_j\}). \]
Example: 1D chain

Reflection (E and Huang 2001, 2002)

\[ R(k) = \frac{1 - \sum_j e^{ijk} \int_0^t \alpha_j(\tau)e^{i\omega\tau} d\tau}{1 - \sum_j e^{-i\omega \tau} \alpha_j(\tau)e^{i\omega\tau} d\tau}. \]
Example: 1D chain

Premature truncation vs variational BC
Example: 2D triangular lattice

the triangular lattice

the first Brillouin zone
Example: 2D triangular lattice

neighbor atoms    phonon reflection for $J = 26, M = 2$

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Example: 2D triangular lattice

size of the stencil

phonon reflection along $\theta = 2\pi/3$

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Boundary conditions for molecular dynamics
Application to fracture simulations

Fixed BC
Application to fracture simulations

Local BC
More practical issues: external loading

stress free?

The reflection coefficient $|R(k)| \equiv 1$!
More practical issues: external loading

Applied deformation:

\[ W = F n - n = \frac{\partial u}{\partial n}, \]

Boundary condition:

\[ w_0 = \sum_j \int_0^{t_0} \alpha_j(s)w_j(t - s)ds. \]

In Fourier space:

\[ \hat{w}(k) = \hat{u}(k)(1 - e^{iak \cdot n}). \]
Summary

1. analysis of phonon reflection
2. variational formulation to minimize phonon reflection
3. **local** boundary condition
4. implementation issues
5. application to fracture simulations