

## Multiscale Simulations of Brittle Fracture and the Quantum-Mechanical Nature of Bonding in Silicon

N. Bernstein and D. Hess

Center for Computational Materials Science, Naval Research Laboratory,  
Washington, DC 20375, USA.

### ABSTRACT

We simulate the microscopic details of brittle fracture in silicon by dynamically coupling empirical-potential molecular dynamics of a strained sample to a quantum-mechanical description of interatomic bonding at the crack tip. Our simulations show brittle fracture at loads comparable to experiment, in contrast with empirical potential simulations that show only ductile crack propagation at much higher loading. While the ductility of the empirical potentials can be attributed to their short range, it is unclear whether the increased range of the tight-binding description is sufficient to explain its brittle behavior. Using the multiscale method we show that at a temperature of 1100 K, but not at 900 K, a dislocation is sometimes nucleated when the crack tip impinges on a vacancy. While this result is too limited in length and time scales to directly correspond to experimental observations, it is suggestive of the experimentally observed brittle to ductile transition.

### INTRODUCTION

Many materials, ranging from silicon to ferritic steels, exhibit a transition between brittle and ductile fracture as a function of temperature [1]. The qualitative differences between these two modes of failure lead to great differences in material toughness. While long-range elastic strain fields, well described by continuum elasticity theory, provide the energy that drives fracture and failure, the nature of fracture itself is ultimately determined on the scale of atoms and the electrons that bind them. Therefore, a microscopic description is required for reliable prediction of the nature of fracture in a given material.

Silicon has become a model system for the brittle to ductile transition (BDT) since it shows a particularly sharp transition between the two types of behavior [1]. Below the BDT temperature (about 850 K) failure occurs at the Griffith criterion [2], when elastic energy relief due to crack elongation balances the energetic cost of the new surface. Atomistic simulations of fracture in silicon using empirical potentials (EP) show a disordered zone around an atomically blunt crack, shown in Fig. 1, which propagates only at much higher loadings than the Griffith criterion [3, 4, 5]. Ab-initio simulations of quasistatic fracture of small samples show brittle behavior with some lattice trapping but no disorder [6, 7]. To explore the nature of fracture in silicon at the atomic scale, we have developed an atomistic simulation technique based on molecular dynamics (MD). The method incorporates a simple quantum-mechanical description of atomic interactions near the crack tip, dynamically coupled to EP MD far from the crack tip. In contrast to simulations

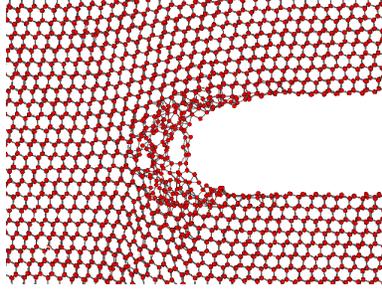


Figure 1: A snapshot from a simulation of fracture using the EDIP empirical potential. The crack tip is blunt, and a disordered region has formed ahead of the crack.

utilizing EP models alone, our results show good agreement with experiment. This suggests that an accurate quantum-mechanical treatment of bonds near the crack tip provides a qualitative improvement in description of fracture over the EP models used previously.

## METHOD

Recognizing that fracture involves physical processes on a wide range of length scales, Abraham *et al.* constructed a multiscale tool for simulating fracture. They dynamically coupled time-dependent finite elements for the far field, EP MD for atoms on the mesoscopic length scales, and tight-binding (TB) MD in a nm sized region that includes the crack tip [8]. Other groups have also presented similar approaches that couple various subsets of these methods [9, 10, 11, 12, 13]. Here, we simulate the process of dynamic fracture in silicon with a coupling of length scales approach using two regions [5]. Far from the crack we use EP MD with the environment dependent interatomic potential (EDIP) [14, 15]. In the immediate vicinity of the crack tip we include explicitly the electrons that mediate interatomic bonds through a quantum-mechanical model based on a semi-empirical TB total-energy method [16]. The tight-binding forces are computed using a Green’s function based method that scales linearly with the number of atoms [5, 17]. The MD time evolution is computed using the velocity Verlet algorithm [18] at constant energy with a 1.0 fs time step. The system consists of a  $3 \times 25 \times 60$  supercell of a 12 atom  $(\frac{1}{2}\bar{1}0) \times (11\bar{1}) \times (\frac{1}{2}\bar{1}\bar{1})$  (from here on denoted as  $x$ ,  $y$ , and  $z$ ) unit cell. Periodic boundary conditions are used along  $x$  and  $z$ . The thin, periodic geometry of the system along  $x$  leads to a nearly two-dimensional plane-strain configuration in the  $y$ - $z$  plane.

A seed crack with  $(11\bar{1})$  faces and a  $\langle 1\bar{1}0 \rangle$  crack front is introduced by removing a bilayer in the the  $x$ - $z$  plane. The crack extends through the entire system along  $x$ , and is 198.6 Å long (half the system size) along  $z$ . The crack is centered with respect to the sample in the  $z$  direction, so that it has two tips, one at each end. The entire system includes over 53000 atoms. We use a TB region that is about 17 Å (five bilayers) thick along  $y$ , and about 45 Å long (17 lattice periodicities) along  $z$ , surrounded by a 6.5 Å coupling layer. This volume, including the coupling layer, includes about 1000 atoms. The TB region is placed around one of the tips of the seed crack, and is not moved during the simulation. Fig. 2 shows the configuration of atoms and the TB region near the crack tip.

The initial configuration is set by moving each atom according to the elastic displacement

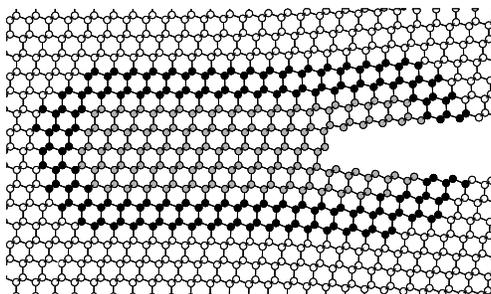


Figure 2: Atomic configuration in the vicinity of one of tips of the seed crack. EP region atoms are drawn in white, TB region atoms in grey, and coupling layer atoms in black.

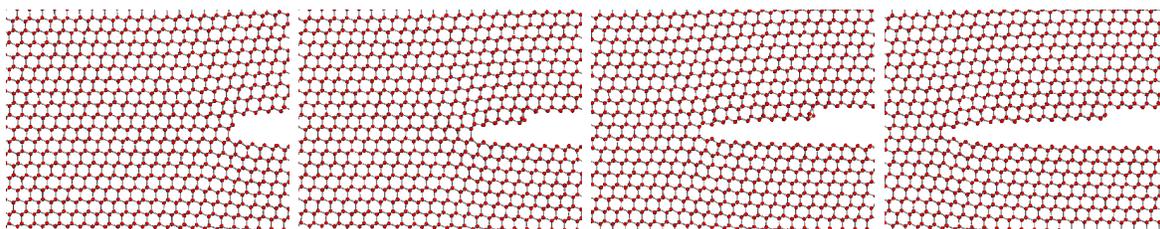


Figure 3: Snapshots from a multiscale simulation at  $T = 200$  K and 1.7% applied strain at 0.4 ps, 1.0 ps, 1.6 ps and 2.1 ps (from left to right).

field in a strained infinite plate with an infinitely thin crack [19], and fixing the  $+y$  and  $-y$  surfaces. We rescale the EDIP length scale (parameter  $c$  of ref. [15]) to reduce the EDIP equilibrium lattice constant to the TB model equilibrium value of  $5.406 \text{ \AA}$ .

## RESULTS

Four snapshots from a simulation of a system under 1.7% strain at  $T = 200$  K are shown in Fig. 3. The crack retains an atomically sharp tip, propagating in a straight line without branching or meandering, and leaves behind a smooth, unreconstructed surface. The velocity of the crack tip as a function of loading is plotted in Fig. 4. Onset of crack propagation is just above the Griffith criterion, in good agreement with experiment. The limiting speed of the crack, about  $2.7 \text{ m/s}$ , is 0.6 times the Rayleigh wave speed of EDIP. The saturation of the crack speed at about 0.6 to 0.9 of the Rayleigh wave speed appears to be universal, having been observed in our simulations using EDIP and Stillinger-Weber (SW), as well as a modified form of SW that gives brittle fracture [3], and experiment [2, 20, 21]. It is interesting to note that this speed is close to the critical speed for instability to branching seen in ball and spring models [21], although we have not observed such an instability.

In experiment, ductile behavior above the BDT temperature is accompanied by a large number of dislocations. To see if our model can shed some light on the microscopics of the BDT, we simulated the fracture of a sample at  $1100 \text{ K}$ . While we observed no dislocations in simulations of fracture in a perfect crystal, a vacancy directly ahead of the crack tip altered this behavior. Snapshots from one such simulation are shown in Fig. 5. Upon reaching the vacancy a  $90^\circ$  shuffle set partial dislocation is nucleated immediately above

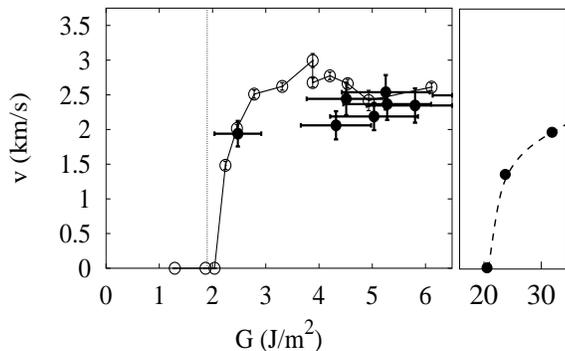


Figure 4: The speed of the crack tip as a function of energy release rate, which is proportional to the applied strain squared. Solid line with open circles is the results of the present work, solid circles with error bars are experimental data from ref. [2], and dashed line is an approximate sketch of EDIP simulation. The vertical line indicates the Griffith criterion for the multiscale simulation.

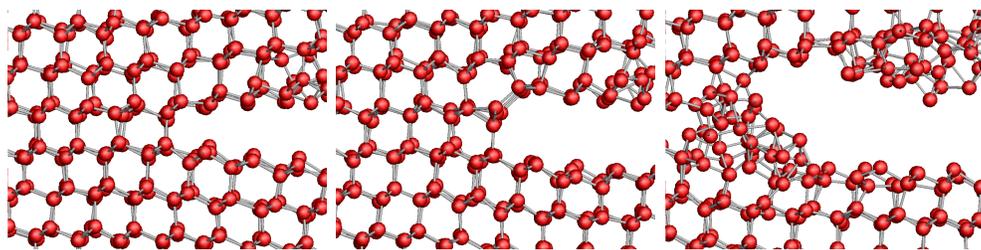


Figure 5: Snapshots from a multiscale simulation at  $T = 1100$  K and 1.7% applied strain of a sample with a vacancy ahead of the crack. The left panel shows the crack tip immediately before reaching the vacancy. The middle panel shows the nucleated dislocation core, visible as a seven-fold ring adjacent to a five-fold ring. The right panel shows the system at the end of the simulation, with the arrested crack tip.

the crack tip, and the crack arrests. Repeating the same simulation 11 times, changing only the initial velocities, has shown this behavior twice. At a temperature of 900 K, no dislocations nucleated in 10 repetitions of the simulation.

## DISCUSSION

The material properties that lead to the difference between the EP and multiscale simulation are not obvious despite the striking contrast in their morphologies. One model, proposed by Rice, provides a quantitative measure of the tendency of a material to be brittle or ductile in terms of the ratio of the energies for surface and dislocation formation  $\gamma_s/\gamma_{us}$  [22]. Values of Rice's criterion for silicon computed using different methods are shown in Table 1. While none of the models agree perfectly with the LDA calculation, which is expected to be quite accurate, no trend is apparent. For both glide and shuffle set dislocations, Rice's criterion for the TB model is quite similar to at least one of the EP models. Therefore application of Rice's criterion would predict that the multiscale simulation should be quite similar to an EP simulation. Since this is not the case, clearly something other than this energetic criterion is controlling the behavior.

One alternative is a criterion based on stresses rather than energies [4]. To describe covalent

Table 1: Rice criterion for glide and shuffle set dislocations computed using LDA, TB, EDIP, and SW.

	LDA	BK-TB	EDIP	SW
$\gamma_s/\gamma_{us}$ (glide)	0.90	0.40	0.59	0.45
$\gamma_s/\gamma_{us}$ (shuffle)	1.02	0.90	0.85	1.71

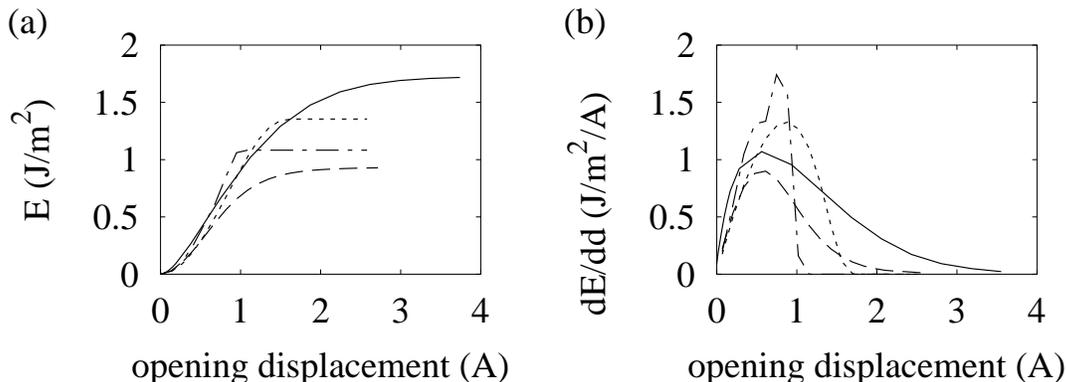


Figure 6: Energy as a function of displacement for two blocks of silicon pulled apart rigidly, exposing a (111) surface (panel a), and the stress, defined as the derivative of the energy (panel b). Solid line is LDA, dashed line is TB, dotted line is SW and dash-dotted line is EDIP.

bonding in a simple functional form, EP models such as SW and EDIP include only nearest neighbor interactions. In TB and LDA, and presumably in experiment, two surfaces interact even when the atoms are substantially farther apart. Since the surface energies of all the models are similar, the stresses needed to pull apart two blocks of silicon to form a new surface are much higher for the short range potentials, as shown in Fig. 6. Therefore, even when it is energetically favorable for the crack to propagate, the stress at the crack tip may not be high enough to break a bond. The stress needed to form a dislocation, on the other hand, is not systematically overestimated by the short range EP models. As a result, for the EP models the shear stress directly above the crack tip exceeds the value needed to nucleate a dislocation before the tensile stress is high enough to break a crack tip bond. While the range of the potential appears to explain why the empirical potentials are ductile, it is not necessarily sufficient to explain why TB is brittle. Another possibility is suggested by the explicitly quantum-mechanical description of the electrons that form the interatomic bonds. This inherently non-local description is only approximated by the three body and coordination dependent terms in the EP models. However, some more subtle non-local effect, for example a weakening of the crack-tip bond by the local strain, could also reduce the effect of the peak stress for the TB model and therefore the multiscale simulation.

## CONCLUSION

We have developed a multiscale simulation tool that allows us to study fracture and other multiscale phenomena with accuracy and efficiency. The dynamical coupling of TB and EP atomistic simulations has produced a simulation of dynamic brittle fracture in silicon that

is in good agreement with experiment. In contrast, EP simulations so far have shown only ductile fracture at unphysically high loads. The quantum-mechanical description of bonding at the crack tip gives the multiscale method the necessary accuracy, while the rest of the system is more efficiently (and sufficiently accurately) described by an EP. The qualitative difference in the morphology of fracture between empirical potentials and the multiscale approach cannot be attributed to Rice's energetic criterion. It seems likely that for the empirical potentials the short range of the interactions magnifies the peak stress that the system can sustain to unphysical levels. However, it is not clear that the intermediate range of the TB model is sufficient to explain its brittleness, and the fundamental reason for the qualitative differences is still an open question. Our observation of dislocation nucleation at 1100 K, but not at 900 K, suggests that a BDT may occur at a temperature close to the experimental value.

## ACKNOWLEDGEMENTS

This work is supported in part by a grant of computer time through the DOD HPC Grand Challenge program at the ASC MSRC and MHPCC DSRC. The authors thank the Office of Naval Research for its support.

## REFERENCES

- [1] P. B. Hirsch and S. G. Roberts, *Phil. Mag. A* **64**, 55 (1991).
- [2] J. A. Hauch *et al.*, *Phys. Rev. Lett.* **82**, 3823 (1999).
- [3] D. Holland and M. Marder, *Phys. Rev. Lett.* **80**, 746 (1998).
- [4] D. Holland and M. Marder, *Adv. Mat.* **11**, 793 (1999).
- [5] F. F. Abraham *et al.*, *Mat. Res. Soc. Bull.* **25**, 27 (2000).
- [6] J. C. H. Spence, Y. M. Huang, and O. Sankey, *Acta Metall. Mater.* **41**, 2815 (1993).
- [7] R. Perez and P. Gumbsch, *Phys. Rev. Lett.* **84**, 5347 (2000).
- [8] F. F. Abraham *et al.*, *Europhys. Lett.* **44**, 783 (1998).
- [9] S. Kohlhoff, P. Gumbsch, and H. F. Fischmeister, *Phil. Mag. A* **64**, 851 (1991).
- [10] W. G. Hoover, A. J. D. Groot, and C. G. Hoover, *Comp. Phys.* **6**, 155 (1992).
- [11] J. A. Nieminen and S. Paavilainen, *Phys. Rev. B* **60**, 2921 (1999).
- [12] W. Cai *et al.*, *Phys. Rev. Lett.* **85**, 3213 (2000).
- [13] G.-H. Wen *et al.*, *Int. J. Quant. Chem.* **78**, 459 (2000).
- [14] M. Z. Bazant, E. Kaxiras, and J. F. Justo, *Phys. Rev. B* **56**, 8542 (1997).
- [15] J. F. Justo *et al.*, *Phys. Rev. B* **58**, 2539 (1998).
- [16] N. Bernstein and E. Kaxiras, *Phys. Rev. B* **56**, 10488 (1997).
- [17] N. Bernstein, In preparation.
- [18] M. Tuckerman, B. J. Berne, and G. J. Martyna, *J. Chem. Phys.* **97**, 1990 (1992).
- [19] K. B. Broberg, in *Cracks and Fracture* (Academic Press, San Diego, 1999), p. 132.
- [20] T. Cramer, A. Wanner, and P. Gumbsch, *Phys. Rev. Lett.* **85**, 788 (2000).
- [21] M. Marder and X. Liu, *Phys. Rev. Lett.* **71**, 2417 (1993).
- [22] J. R. Rice, *J. Mech. Phys. Solids* **40**, 239 (1992).