Ab Initio Theory of Dislocation Interactions: From Close-Range Spontaneous Annihilation to the Long-Range Continuum Limit

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Parallel supercomputer technology now permits *ab initio* studies of systems of sufficient size to explore the interactions among dislocations in a solid. This study shows that the silicon shuffle-set (110) screw dislocation is stable against spontaneous dissociation, provides an *ab initio* value for the dislocation core energy, demonstrates a dislocation-antidislocation interaction approaching the classical limit *within a few tens of angstroms,* and reveals a pathway for the spontaneous mutual annihilation of a dislocation dipole of the type that occurs when a Frank-Read source emits a dislocation loop.

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One dimensional topological defects play a fundamental role in a variety of areas of condensed matter physics from flux lines in type II superconductivity to Kosterlitz-Thouless vortex-pair excitations in *two* dimensional phase transitions [1] to vortex lines in inviscid fluid flow to dislocations in solids. Because of short-wavelength logarithmic divergences, theoretical treatments of the energetics of these defects often introduce an *ad hoc* cutoff at some microscopic length scale. In this Letter, we present calculations that overcome this "ultraviolet catastrophe" in a realistic system and that therefore provide reliable microscopic information such as the dislocation core energy. To do this, we treat the short-wavelength physics properly through a first principles quantum mechanical approach while studying a system of sufficient size to exhibit macroscopic behavior. The local-density, nonlocal pseudopotential, supercell framework [2] provides the best compromise between reliability and tractability for such an application, and as that approach is most conveniently applied to periodic systems, we shall focus on the physics of dislocations in solids.

In the solid state, it is dislocations, primarily, that are responsible for the mechanical properties of macroscopic matter. They also play an important role in semiconductor technology as well, where the performance of devices is degraded by the conduction pathways which dislocations provide [3] as they form at elevated temperatures either during fabrication [4] or operation [5]. Understanding dislocation processes requires knowledge not only of the basic properties of the dislocations themselves but also of the mechanisms of their creation and propagation. These mechanisms, in turn, are controlled by the interaction of dislocations with point defects such as impurities, the interaction of dislocations with other extended defects such as surfaces and grain boundaries, and the interactions of dislocations among themselves [6].

The study of dislocations has developed successfully over the last sixty years as a classical continuum theory. However, continuum theory cannot describe the atomic processes that occur as two dislocation cores come into contact nor give *a priori* information on the limits of its own applicability at short length scales. Atomistic effects in dislocations of diamondlike materials were first considered by Shockley [7], Read [8], and later Hornstra [9], who all reasoned qualitatively from the principle that diamondlike materials prefer to maintain local tetrahedral coordination. Later studies used semiempirical classical potential methods to determine quantitative structures, usually combining the resulting coordinates with semiempirical quantum calculations to provide electronic information as well [10–16]. It is only within the last year or two that supercomputer technology has developed sufficiently (primarily through the introduction of parallel processing) to allow more reliable, parameter-free *ab initio* quantum treatment of dislocation systems. *Ab initio* dislocation studies have so far focused either on the core structure itself [17] or on the effects of impurities on the core structure [18], but have yet to address the issue of the interactions among dislocations.

In this Letter we study specifically the interactions among dislocations on the technologically important {111} slip system of silicon. To avoid the complexity of comparing the energies of systems with differing amounts of material, we consider pure screw dislocations, which have ⟨110⟩ Burgers vectors on the {111} slip system. Associated with the two atom basis of the silicon lattice are two distinct microscopic geometries for the ⟨110⟩ screw dislocation, commonly referred to as the "glide" and the "shuffle" sets [6]. Experimental observation of plastically deformed samples reveals ⟨110⟩ screw dislocations of the so-called "glide" set [19] dissociated into partial dislocations bounding stacking faults with widths on the order 30–50 Å [20]. First principles study of the interaction between even a pair of dislocations of such extent would be prohibitive even for the latest generation of massively parallel supercomputers. In contrast, the splitting of *shuffle-set* dislocations into partials is a more complex process, leaving structures with geometries which, unlike the basic 30° and 90° partial dislocations of the glide set [15,17,21], are not suited to reconstructions which restore the fourfold coordination of the lattice [9]. Accordingly, one may expect the shuffle ⟨110⟩ screw dislocation to be stable against spontaneous

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dissociation into partials (as confirmed by our calculations) and thus be suited to this *initial* first principles quantum treatment of dislocation interactions.

For this study, we calculated (within the aforementioned density-functional framework [2]) the energies of periodic supercells containing dislocation dipoles at differing separations (R = 3.3, 10, 16, 23 Å). The basis set for expanding the electronic wave function includes plane waves up to a cutoff of 6 Ry [22], and two special k points sample the Brillouin integrations. To simplify comparison, all dislocation dipoles are embedded in orthorhombic supercells of the same size, containing 324 atoms and 59 Å × 28 Å in extent in the (110) plane perpendicular to the screw axis. (Figure 1 shows the basic geometry of the calculation.) To produce the atomic configurations, we first set the bonding *topology* according to Hornstra's proposed tight-core shuffle configuration [9] for a screw dislocation-antidislocation pair separated along the $[\bar{1}12]$ direction and then find a suitable *geometry* by relaxing the lattice within a simple Keating model where the bonding topology can be enforced as a constraint [23]. From this initial geometry, we then relax the atomic coordinates under the influence of the electronic wave functions in the full *ab initio* calculation until the lattice energy converges to within less than one-tenth of an electron volt per supercell.

Figure 2 displays the final relaxed structure of a dislocation core taken from the maximally separated configuration of our study (23 Å). All atoms in this *ab initio* structure remain fourfold coordinated, and the Hornstra tight-core topology is stable despite significant local strain in the lattice, where bonds are stretched and bent by as much as 9.2% and 23°, respectively.

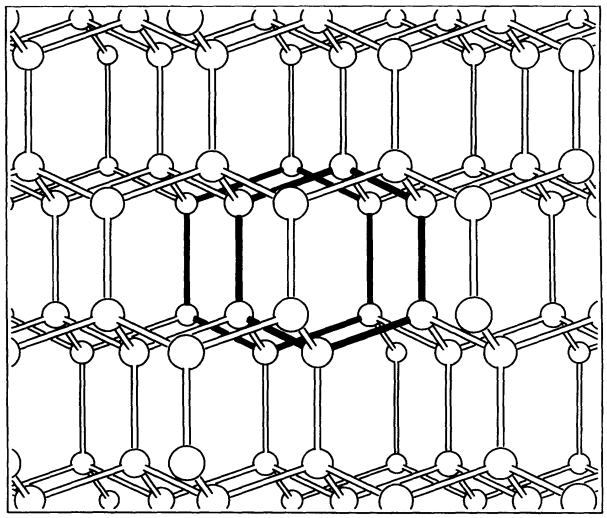

FIG. 1. Three dimensional perspective view of bulk silicon in the geometry of the present calculation. The $[1\bar{1}1]$ direction runs vertically, and, apart from a 16° rotation to the right to facilitate viewing, the [110] screw axis runs perpendicularly out of the page and the $[\bar{1}12]$ direction runs horizontally from left to right. The supercell described in the text extends 28 Å to the right, 59 Å vertically, and contains a total of 324 atoms. Black bonds indicate two sixfold rings stacked along the [110] axis.

As the distance between defects in our calculation decreases, the local strain steadily increases as the strain fields from the two defects overlap until the two defects are centered on neighboring columns of sixfold rings (Fig. 3). At this point, the silicon bonding network no longer can support the microscopic stress, and the initial Keating-refined Hornstra configuration relaxes steadily through the transition state of Fig. 4 to the bulk silicon lattice configuration *without crossing an energy barrier.* In this *spontaneous* annihilation process, the bonds crossing the plane connecting the two dislocation lines yield to the stresses pulling the atoms involved in these bonds toward their bulk locations to the extent that these bonds break and the atoms eventually approach and bond with their partners in the bulk configuration instead. A Frank-Read source [24] tied to a dislocation loop segment by a pair of dislocation lines in this geometry will emit a full dislocation loop into the crystal as the lines migrate under external strain into *neighboring* columns of sixfold rings and annihilate along the pathway which Fig. 4 illustrates.

While the relatively large dimensions of our supercell combined with the decay of strain fields away from a dislocation ensure a good representation of the core structure of the isolated dislocation in Fig. 2 and the dislocation dipole annihilation process depicted in Fig. 4, the logarithmic *growth* with distance of the interaction energy of two dislocations demands much more delicate consideration of the effects of periodic boundary conditions on the energy of our supercell. The excitation energy (per unit length along the dislocation axis) of our dipole supercell relative to bulk crystal differs in *two* ways from the energy which interests us, the energy of an isolated dipole at finite separation, $E_{\text{dipole}}(R)$. First, by holding the lattice vectors of

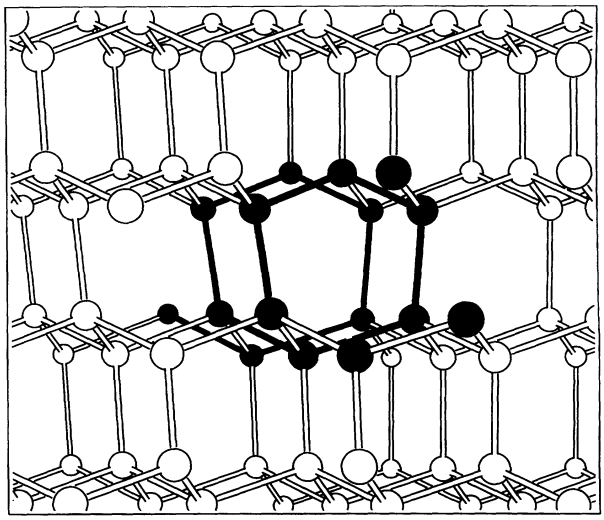

FIG. 2. Stable *ab initio* core structure for an isolated compact shuffle ⟨110⟩ screw dislocation in the Hornstra tight core shuffle topology (same view as Fig. 1). Black atoms outline the core of the defect, and black bonds indicate two microscopic Burgers circuits about the dislocation core that run clockwise into the page.

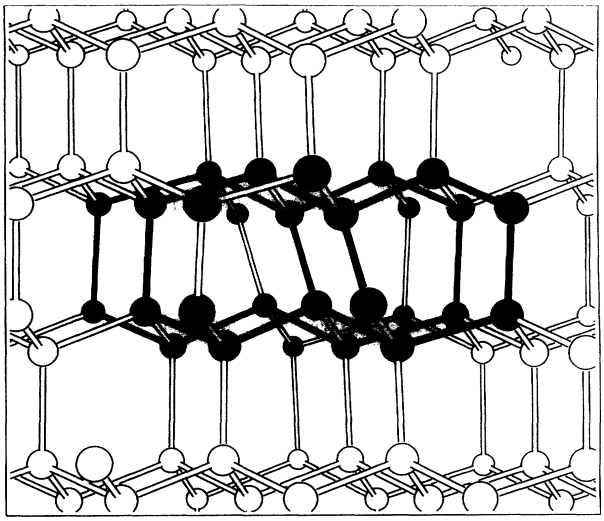

FIG. 3. Relaxed Keating configuration for a proximate dislocation dipole in the Hornstra topology separated 3.3 Å along the $[\bar{1}12]$ direction. Black atoms make up the cores of the two component dislocations. (The dislocation on the right corresponds to the defect in Fig. 2.) Black bonds of the dislocation on the right (left) comprise two microscopic Burgers circuits running clockwise into (out of) the page. Separating the component dislocations further from one another generates additional rows of tilted vertical bonds between the dislocation cores.

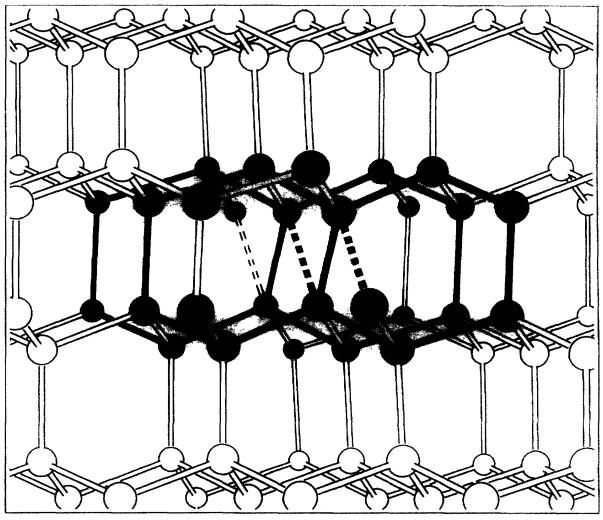

FIG. 4. Transition state through which the dipole in Fig. 3 passes during spontaneous annihilation. As the titled vertical bonds of the dipole (now dashed) yield to the stress, the atoms in the center of the defect move toward and eventually bond (new solid lines) with their bulk partners to reform the closed sixfold rings of the crystal.

the unit cell fixed, we have imposed a stress not present in the periodic dislocation dipole array because, as Bigger *et al.* [17] point out, the effect of such an array is to introduce a bulk shear strain into the crystal. The energy associated with this effect is easily calculated by computing the energy released within the Keating model, which is well suited and parametrized to describe such bulk elastic effects, upon relaxation of the orthorhombic boundary conditions. Once we have the correct excitation energy per unit cell for the fully relaxed dislocation dipole lattice, ΔE_{cell}, the second effect we must consider is the energy contribution from the periodic images of the dislocation dipole in our cell. If the supercell is sufficiently large (as we verify below), continuum theory then correctly predicts the size of this effect, splitting the energy of the dislocation dipole lattice into $E_{\text{dipole}}(R)$ and a Madelung-like term,

$$\Delta E_{\text{cell}} = E_{\text{dipole}}(R) + \left(\frac{\mu b^2}{2\pi} \sum_{\vec{L}\neq 0} \ln \frac{\|\vec{L} - \vec{R}\| \cdot \|\vec{L} + \vec{R}\|}{L^2} \right), \quad (1)$$

where b^2 is the square of the Burgers vector of the component dislocations, $\vec{L}$ ranges over the vectors of the dipole lattice, and μ is an appropriate combination of anisotropic elastic constants for $\langle 110 \rangle$ screw dislocations in the silicon lattice [6]. Note that the net zero Burgers vector of each dislocation dipole ensures that the dislocation core energy does not appear in the Madelung term. Classical continuum theory further gives $E_{\text{dipole}}(R)$ in terms of the energy contained in the lattice beyond some arbitrary (macroscopic) distance r_c from the centers of the dislocations (where the distortions are mild) *and* the classically undetermined, albeit physical, lattice excitation energy contained within a distance r_c of an isolated dislocation core, $E_c(r_c)$,

$$E_{\text{dipole}}(R) = 2E_c(r_c) + \frac{\mu b^2}{2\pi} \ln \frac{R}{r_c}. \quad (2)$$

A least-squares fit of $\Delta E_{\text{cell}}(R)$ from our stress-corrected microscopic calculations for the three stable configurations in our study to Eqs. (1) and (2) gives the value $\mu b^2/2\pi = 0.69 \pm 0.26$ eV/Å for our *ab initio* silicon lattice, consistent with the value 0.92 eV/Å determined from elastic theory and the experimentally determined elastic constants [25]. Armed with this value of μ, Eq. (1) then relates our calculated energies to $E_{\text{dipole}}(R)$. Figure 5 displays the resulting *ab initio* prediction of the energy of an isolated dislocation-antidislocation pair. Note that, apart from not predicting the spontaneous annihilation of the proximate core-anticore pair, the continuum form describes the lattice distortion energy extremely well, even down to the preannihilation energy of the dipole at the *smallest* microscopic separation (to which the curve was *not* fitted). The interaction we calculate exhibits the classical limit within just *a few tens of angstroms*, justifying our classical treatment of the periodic supercell images and allowing us to extract from our fit a reliable *ab initio* value for the core energy of the shuffle-set (110) screw dislocation. At our best fit value for μ, we find the core energy, referenced at the Burgers vector, to be $E_c(r_c \equiv b) = 0.56 \mp 0.21$ eV/Å. (Fixing μ at values above the optimal $\frac{\mu b^2}{2\pi} = 0.69$ results in *lower* values of E_c to make the best fit.)

As a point of reference, the energies of our initial configurations in the Keating potential may be put through the same analysis to yield a classical atomistic estimate for the core energy. The Keating potential is a simple expansion about the perfect crystal and should not be

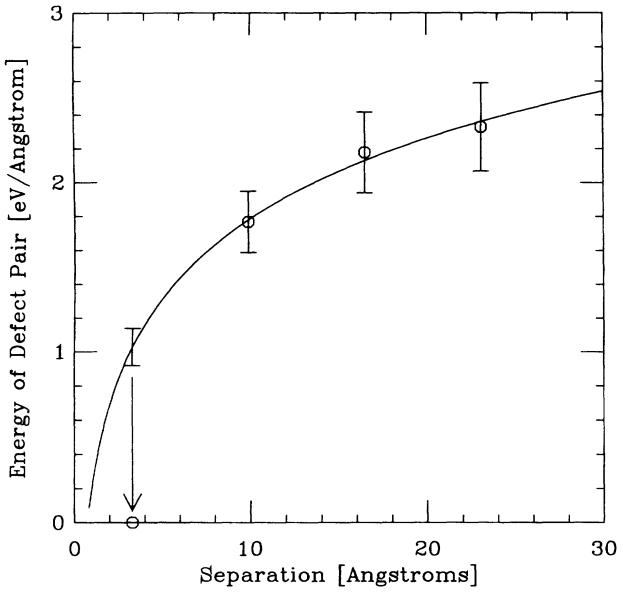

FIG. 5. *Ab initio* prediction for the energy (relative to bulk) of an isolated dislocation-antidislocation pair (points with ±10% error bars; see 17) and the elastic continuum theory form (curve) for the same quantity. The arrow indicates the spontaneous annihilation of the defect of Figs. 3 and 4 back to the bulk configuration. The preannihilation energy of this configuration is the *ab initio* energy of the configuration of Fig. 3 with a small, ≈ 0.1 eV/Å, long-range relaxation energy estimated from the magnitude of the preannihilation *ab initio* forces.

trusted *a priori* in the highly strained regions that are critical to the core energy and where, for example, this atomistic model fails to predict the annihilation pathway of Fig. 4. Nonetheless, for the shuffle-set (110) screw dislocation, the Keating model gives results comparable to those from the *ab initio* calculation, overestimating the dislocation interaction strength, $\frac{\mu b^2}{2\pi} = 1.10$, and yielding a similar value for the core energy, $E_c(b) = 0.72$.

The tetrahedral bonding network of the dislocation core in this study is most likely responsible for this system's rapid approach to the continuum limit. The knowledge that the continuum limit is approached so rapidly in fourfold coordinated silicon systems, even when containing highly strained bonds, is of significant practical importance because the component partial dislocations of the experimentally observed *glide* set most likely reconstruct to tetrahedral configurations [15,17,21]. Our results suggest that quality energetics for such systems may be obtained in relatively small cells by compensating for the effects of periodic images within elastic continuum theory. It may now be possible, for instance, to extract core energies for *extended* glide set dislocations from feasible, or already existing, calculations of the energetics of the separate components of these dislocations. Future calculations in larger supercells will allow us to gauge the presence of any renormalization of the bulk elastic constants at small length scales, and, finally, the core energy extraction procedure we employed above may be used to determine the effects of the presence of impurities and defects on the core energies of dislocations and thus open the process of dislocation pinning to *ab initio* study.

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