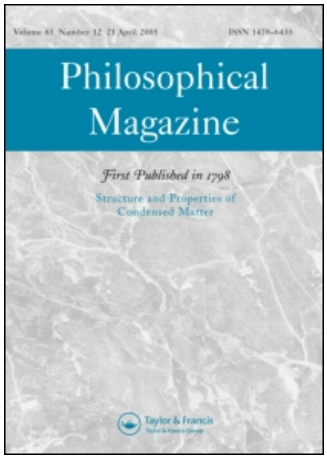


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# Calculations of dislocation mobility using Nudged Elastic Band method and first principles DFT calculations

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# Calculations of dislocation mobility using Nudged Elastic Band method and first principles DFT calculations

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We present a new technique which makes it possible to determine mobility properties of dislocations with first principles accuracy without having to apply corrections for the influence of boundary conditions. The Nudged Elastic Band method is used together with periodic boundary conditions and all dislocations included in the simulated cell are coherently displaced during the calculations. The method is applied to the displacement of a non-dissociated shuffle screw dislocation in silicon along two different directions. Peierls energies as well as dislocation structure as a function of the dislocation position in the lattice have been obtained. We have determined the Peierls stresses for both directions, in excellent agreement with previous determinations. Finally, we discuss the advantages of the technique over other methods.

## 1. Introduction

Dislocations modelling has first been developed in the framework of linear elasticity theory, not only because of historical reasons, but also because this approach has been able to capture most of the dislocations physical properties [1]. However, this approach is inadequate to deal with the severe lattice distortion near the core of a dislocation. In consequence, it is not suited for describing important properties governed by the structure of the core, such as dislocation mobility or dislocation interactions at short distances. Major improvements have been obtained thanks to the Peierls–Nabarro (PN) model and its recent developments [2–4], since it combines elasticity theory with the discrete nature of the lattice. This model removes the artificial divergence at the core originating from the continuum description, allowing us to determine several key quantities such as the core width, the Peierls energy and the Peierls stress. Nevertheless, while this approach has been proved to be successful in many cases, it is not accurate enough for several classes of materials such as covalent or ionic materials, in which dislocation cores are very narrow or reconstructed. Although the PN model has been recently improved [4], it intrinsically includes some limitations by construction [5].

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A full atomistic treatment of the dislocation is expected to yield better results. Hence, accurate values can be determined in case of fcc metals, for which reliable classical interatomic potentials are available. However, for other materials such as semiconductors or ceramics, energetic properties of dislocation cores are poorly described by potentials. This point has been brought to light in the case of silicon for example [6]. Another option is to use first principles density functional theory (DFT) calculations which give an accurate description of most materials even those including unusual and unfavourable bonding configurations. The problem with these methods, however, is that the simulated systems are limited in size, and as a consequence boundary conditions require careful and special treatment [7–9], or the use of multiscale modelling methods [10, 11]. This is especially true for Peierls stress determination. Recently, Pizzagalli and Beauchamp have calculated the Peierls stress of screw dislocation in silicon using reduced systems and both periodic and fixed conditions [12]. This work shows that Peierls stress determination using first principles accuracy is possible, but that corrections are required to compensate for boundary effects.

In this paper, we propose a method allowing an accurate determination of the Peierls stress, without the need for boundary corrections. Also, the calculations provide the full energy curve for the movement of the dislocation through the lattice, i.e. the whole Peierls barrier, as well as the associated core structure configurations. The method is based on the combination of the Nudged Elastic Band (NEB) method for finding a minimum energy path (MEP) [13] of a transition and first principles density functional theory (DFT) calculations of the atomic forces subject to periodic boundary conditions. Previously, the NEB method has been used in a few studies of dislocation mobility but only in combination with empirical potentials and fixed atom boundaries in finite systems [14–17]. By using periodic boundary conditions, as we do here, smaller systems can be used in the calculation and it becomes easier to make use of first principles DFT methods. In the following we first describe errors coming from fixed boundaries in small system, and we show how this effect is greatly reduced using NEB and periodic boundaries. Then, the Peierls energy and stress are determined with first principles accuracy in the case of a screw dislocation in silicon, moving along two different directions. Finally we discuss the advantages and the limitations of this method, compared to previous models.

The straightforward approach to computational studies of dislocations is to construct a system that contains one dislocation and fixed atoms at the boundaries of the system. In that case, boundary conditions will introduce errors in the energy variation as a function of dislocation position, especially in the case of small systems. This is schematically depicted in figure 1. The initial and final configuration states include the dislocation–surface interaction energy $E_f^1$, positive (negative) for a fixed (free) surface. For the transition state, ideally located in the centre of the system, the interaction energy $E_f^2$ is different, i.e. $E_f^2 \neq E_f^1$. As a result, the calculated curve will be different from the real one (figure 1). For a screw, the energy change $\Delta E$ due to fixed cylindrical boundaries is proportional to the square of the ratio between the dislocation distance from the cell centre and the cell size, with a factor depending on the shear modulus and the anisotropy of the material [18]. For silicon and cluster sizes considered in first principles DFT calculations, $\Delta E$ is about 50 meV. This effect could be reduced by considering larger cluster systems [17].

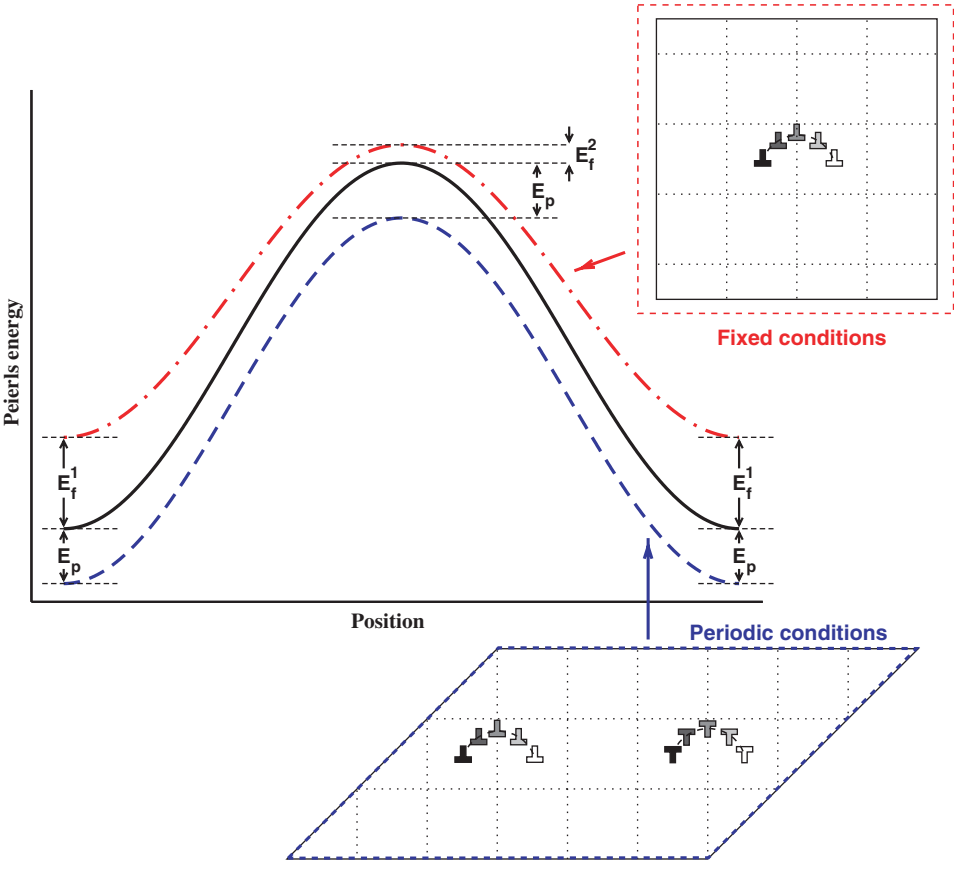


Figure 1. (Colour online) Effect of boundary conditions on Peierls energy variation as a function of the dislocation core position. For fixed conditions, the interaction between the single dislocation and the surface cluster ranges from $E_f^1$ to $E_f^2$ (dash-dotted line). By combining periodic conditions and NEB, and by displacing the dislocations coherently, the interaction $E_p$ between dislocations remains constant (dashed line) and does not affect the deduced Peierls stress and activation energy.

Here, we propose to use periodic boundary conditions, which is well suited for standard plane-waves first principles DFT calculations, and as such have been widely used for investigating dislocations [19–24]. The initial configuration is a system including an even number of dislocations, with a zero Burgers vectors sum (figure 1). The final configuration is then obtained by a simple translation equal to the investigated dislocation displacement. Finally, a standard NEB calculation between these two configurations is performed. The important point here is that the distance between dislocations remains constant along the whole path. This condition is fulfilled at the beginning, if the initial MEP is built from linear interpolation between the end point configurations. We have found that if the springs linking adjacent replicas in the NEB are chosen to be stiff enough and a direct minimization method is used to relax the system to the MEP closest to the initial guess, then the

distance remains constant during the full relaxation until convergence has been reached. The advantage of the method is illustrated in figure 1. Since distances between all dislocations remain constant throughout the NEB calculation, the energy variation as a function of position is simply shifted by a constant term, provided that the total interaction between dislocations $E_p$ is elastic. As a result, the shape of the curve is the same as the real one.

## 2. Model and methods

We apply this method to a non-dissociated screw dislocation in silicon, with an initial stable shuffle configuration [6]. This choice has two motivations. First, in a previous study, the Peierls stress has been determined with first principles DFT and corrections due to periodic and fixed boundary conditions [12]. Second, in the same work, it has been shown that interatomic potentials for silicon are not reliable enough for this particular problem. We then have an interesting system, for which first principles accuracy is required, and with available data for comparison. We considered a system similar to that used in [12], including a dipole of dislocations, a cell geometry of $12 \times 12 \times 1$ (144 atoms) corresponding to an infinite quadrupolar arrangement [12]. First principles DFT calculations have been performed with the VASP distribution [25, 26], with two special k-points along the dislocation line direction, an ultrasoft pseudopotential for Si [27], a plane waves energy cutoff of 140 eV, and the GGA functional PW91 [28]. System relaxation is stopped when all forces are lower than $3 \times 10^{-3}$ eV Å$^{-1}$. With these parameters, a lattice constant of 5.475 Å and a bulk modulus of 99.7 GPa have been obtained, in good agreement with experiments [29]. The NEB calculations have been performed with both improved tangent and climbing images algorithms [30, 31]. Figure 2 shows the zinc-blende structure, and two dislocation displacements, investigated in this work. D′ corresponds to the easiest path for displacing a non-dissociated screw, while D″ is the direct path allowing the screw to cross-slip from shuffle to glide {111} planes.

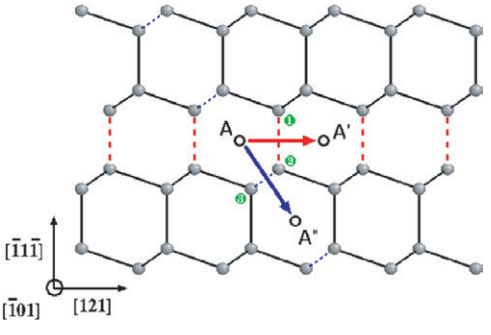


Figure 2. (Colour online) Ball-and-stick representation of the zinc-blende structure (($\bar{1}$01) plane). The path D′ (D″) is defined by a dislocation displacement from A to A′ (A to A″). Dashed lines show bonds linking atoms used for calculating the disregistry due to the dislocation core. In particular, atoms located in dislocation cores for saddle configurations are marked by numbers (1 and 2 for D′, and 2 and 3 for D″).

In order to obtain the Peierls stress from the energy variation, an accurate determination of the dislocation position relative to the lattice is required. Distances between images computed in configuration space, as output by NEB, are useless in that case because the dislocation position does not linearly follow the configuration space coordinates. As stated in [32], dislocation centres cannot be derived unequivocally from the atomic positions. Dislocation centres are then determined according to a well-known procedure. Relative displacements of atoms on both sides of the plane perpendicular to $\{\bar{1}01\}$ and including the dislocation displacement vector are first calculated. Figure 2 shows the bonds linking these atoms for D′ and D″. Then, the dislocation centres are determined by fitting relative displacements using the elastic expression $b/2\pi$ , $\arctan((x - x_0)/\Delta)$, $\Delta$ allowing us to define a dislocation core width [1]. This approach assumes that the whole dislocation can still be described as a Peierls dislocation throughout its displacement in the lattice. As a check, the distance between the two dislocations in the dipole has been determined for all relaxed NEB images. The maximum deviation is $3 \times 10^{-2}$ Å, i.e. less than 1% of the total displacement. This shows that the two dislocations have been coherently displaced, as assumed.

## 3. Results

The Peierls energy, i.e. the energy variation associated with the displacement of a dislocation in a crystalline lattice, is represented in figure 3 for two paths, D′ and D″. Note that a multiplying factor of 1/2 has been applied since two dislocations are included in our cell. For both paths, starting from the stable configuration, the energy is smoothly increasing until a halfway maximum, then decreasing to the next stable configuration. In the initial configuration A, the bond linking atoms 1 and 2 is markedly stretched (figures 2 and 4). When the dislocation moves along D′, this stretching intensifies until the bond breaks at the saddle configuration (figure 4), which is a high symmetry position at the intersection of shuffle and glide planes. A simple force relaxation of this configuration indicates it is metastable. In previous, DFT/LDA calculations, we have found that this configuration was weakly stable [6], with an energy difference of 0.32 eV per Burgers vector, in good agreement with the value of 0.41 eV/$b$ obtained in this work. Finally, atoms 1 and 2 form bonds with neighbour atoms to displace the dislocation core in A′. The D′ migration is then done by the breaking and reforming of a stretched bond, explaining why this mechanism is the easiest. For D″, we found that the migration mechanism is totally different. Initially, the bond between atoms 2 and 3 is slightly stretched and out of the (101) plane (figures 2 and 4). This bond is progressively twisted when the dislocation moves along D″, until it belongs to the (101) plane at the saddle point (figure 4). This corresponds to a screw in the glide set, with a $sp^2$ hybridized core [6]. Actually, it is rather a weakly stable configuration, not seen here due to our limited NEB images number. We then have two totally different mechanisms: in one case, the dislocation is displaced via bond breaking and reforming in one case (D′); and via severe bond twisting in the other (D″). It is also interesting to compare calculated Peierls energies with classical models. In fact, it is common to use a phenomenological expression $W\sin^2(x)$ for approximating the Peierls energy [1]. Introducing our

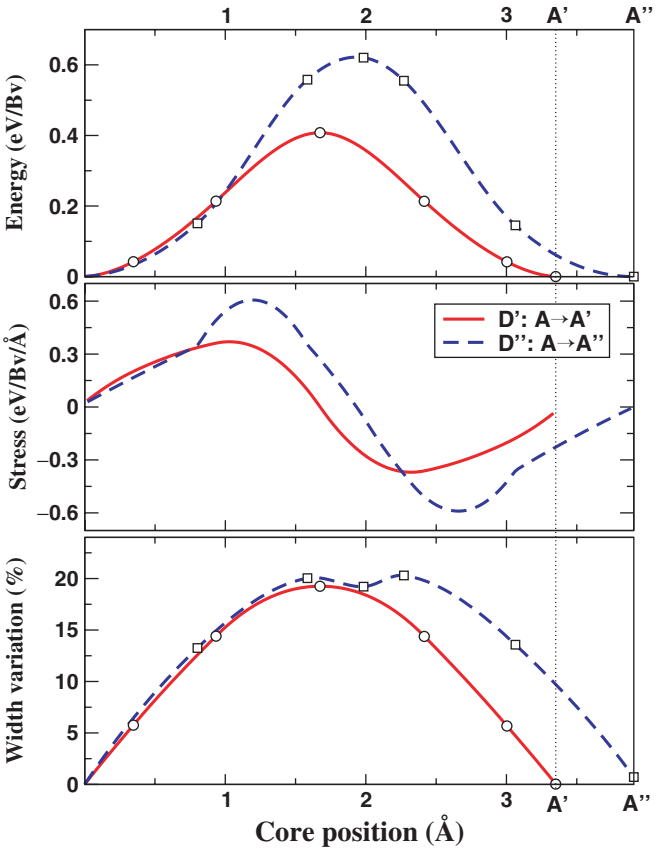


Figure 3. (Colour online) Energy (in eV/Burgers vector, top graph), stress (derivative of the plotted energy curves, middle graph), and core width variation (bottom graph) as a function of the dislocation core position in the lattice for paths D′ (full lines) and D″ (dashed lines).

calculated maximum Peierls energy in this simple model, we obtain a correct estimation of our computed energy curves, in particular for the D′ path.

The Peierls stresses can be accurately obtained as the extrema of the derivative of the spline-fitted energy variations (figure 3), divided by $b^2$, $b$ being the Burgers vector. For the D′ path, the Peierls stress is $2.47\,\mathrm{eV\,Å^{-3}}$, in excellent agreement with the value $2.6 \pm 0.2\,\mathrm{eV\,Å^{-3}}$, obtained from DFT calculations of sheared systems combined with boundary corrections [12]. For the D″ path, the calculated value is $4.05\,\mathrm{eV\,Å^{-3}}$. Obviously, comparing energy barriers and Peierls stresses shows that dislocation displacement would be easiest along the D′ path, so that the dislocation remains in the shuffle plane. Shuffle-glide transition by dislocation cross-slip along the D″ path is then unlikely. This is also in agreement with previous calculations showing that even if a shear stress favouring the D″ displacement is applied on the system, the dislocation is always displaced along the D′ path [12].

Figure 3 also represents variation of the core width $\Delta$ as a function of the dislocation displacement in the lattice. In both cases D′ and D″, the core is

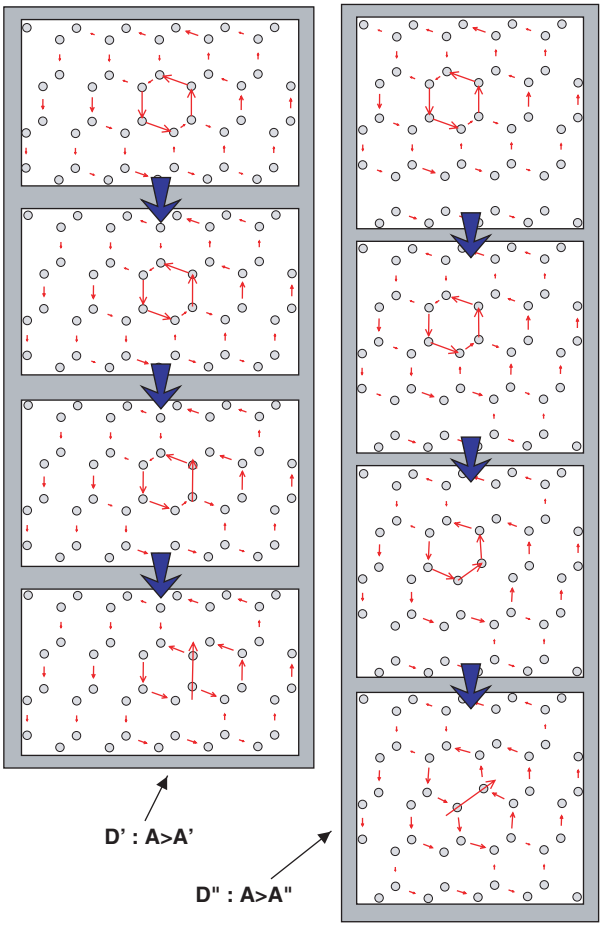


Figure 4. (Colour online) Differential displacement maps corresponding to screw dislocation displacements along D′ (left) and D″ (right), between A and the saddle configurations. The arrows are proportional to the out-of-plane [$\bar{1}$01] shifts between neighbouring atoms introduced by the dislocation.

widened, with a maximum increase of about 20%, corresponding approximately to the maximum Peierls energy. The D″ curve shows a local minimum compared to the D′ curve. A possible explanation is related to the weak stability of the transition configuration along the D″ path, for which the dislocation core is slightly narrowed, whereas for D′, the transition configuration is a true saddle point.

## 4. Concluding discussion

Our approach combining NEB and first principles DFT calculations allows us to determine the energy of a dislocation translated in a crystalline lattice, as well as the associated Peierls stress, without the need for any corrections. In particular, the

calculated Peierls stress for a screw dislocation in silicon is in excellent agreement with published results [12]. In addition to the Peierls energy determination, our approach has several other advantages compared to previous models. In particular, we have access to the dislocation structure during its displacement. Another advantage is the possibility of easily investigating several possible paths, such as the D″ path corresponding to a shuffle-glide cross-slip. We have shown recently that the latter could not be obtained by simply stressing the system [12]. Also, it has been shown recently that the more stable structure of a non-dissociated screw dislocation in the glide set exhibits a $2 \times 1$ reconstruction along the dislocation line [33]. Using our approach, it should be possible to investigate a transition between the single-period shuffle to the double-period glide screw dislocation, provided that the initial system dimension along the dislocation line is $2b$, and that the reconstruction path is simple enough to be obtained with a small number of images in the NEB band.

In our approach, what is really calculated is the coherent displacement of a periodic arrangement of dislocations in a crystalline lattice. One may wonder whether this situation is equivalent to the displacement of a single dislocation. Obviously, even if the distance between dislocations remains constant, the core structure, and then the dislocation core field, is varying during the displacement. So the interaction between dislocations is not constant during the displacement. However, this effect is expected to be negligible and lower than surface–dislocation interactions present in fixed boundary calculations, even in small cell. This is the same assumption done in standard calculations when comparing the stability of several dislocation core configurations, where a constant elastic interactions is assumed between dislocations, independently of the core. In order to get an estimate of this effect, we have performed dislocation calculations with classical potentials and periodic boundary conditions, comparing energy differences between two core configurations as a function of cell size. In all cases, we have found that for the cell size considered here, i.e. $12 \times 12$, the core field interaction energy is lower than 1% of core energy differences. The error is then in the range of standard first principles accuracy, and can be safely neglected.

Another question concerns the fact that during the NEB calculations the coherency of the collective displacement is retained. In fact, it is likely that it is energetically more favourable to first displace one dislocation in the dipole system, then the other. This is especially true in hard materials with covalent bonds for which a strong interaction is expected between close dislocations. However, we usually found in our calculations for silicon that the dislocations were coherently displaced. The key explanation lies in the fact that a relaxation algorithm such as steepest descent or the velocity Verlet algorithm with damped dynamics [13] brings the NEB to the minimum energy path that is nearest to the initial guess even though this may not be the global minimum energy path. While such convergence to the nearest local minimum rather than the global minimum is often a problem when dealing with systems represented by a complex energy surface, it is an advantage here because one can select out the right minimum energy path by choosing the initial path correctly. In order to keep the coherency, it is important that initial and final configurations in the band are well relaxed, with symmetric dislocation cores. We found that if this condition is not fulfilled, a non-coherent displacement could occur.

It is also better to use systems including two dislocations instead of four, since it is easier to keep the coherency in the former case. In NEB calculations, we used standard spring constants (i.e. 5.0 eV $\text{Å}^{-2}$). It is possible that using slightly stiffer springs would also help for keeping coherent displacements.

In conclusion, we propose a new method for investigating the mobility of dislocations using periodic boundary conditions and first principles accuracy. With this method, the Peierls energy and stress, and the dislocation structure during the displacement could be investigated, as well as high energy barriers, out of reach of standard calculations. In principle, the approach could be used for all kinds of dislocations and for all materials, as long as dislocations in the computational cell are stable towards each other.

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