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Stacking fault energies in aluminium

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Abstract. The twin, intrinsic and extrinsic stacking fault energies together with the FCC–HCP structural energy difference are calculated for Al by means of the total energy pseudopotential method. The influence of supercell geometry is controlled by extrapolating the calculated data to infinite cell size. All calculations include full inter-planar relaxations and the final inter-planar separations are presented and shown to vary systematically for the three stacking faults. The calculated stacking fault energies are shown to be consistent with a simple two-parameter model which describes the effective interactions between atomic planes.

1. Introduction

Developments over the last decade of computational methods in theoretical solid state physics together with the continued improvements in computer performance have made it possible to calculate a wide range of material properties. This may lead to a fruitful interplay between experimental and computational methods where the theoretical modelling of material properties can be based not only on experimental input but also on information from *ab initio* calculations on idealized systems.

The stacking faults of crystals play an important role in materials due to their interaction with dislocations. Dislocations often split into partial dislocations with the formation of a stacking fault connecting the partials. The stacking fault region modifies the properties of the dislocation and is therefore important for the understanding of properties like dislocation mobility. An important input to the modelling of dislocations is therefore the relevant stacking fault energies.

Stacking fault energies are tiny—of the order 50 meV per interface atom—and they therefore also constitute a rigorous test case for total energy evaluations. First of all, it is of course important to establish some experience and understanding of the reliability of the local-density-functional scheme which is at the basis of most electronic structure codes today. The local-density approximation aside, the different approaches to total energy calculations contain many other approximations involving the shape of the ionic potential, the basis set used to expand the wavefunctions and the choice of atomic cells. It is therefore also important to compare different total energy approaches and the stacking fault energies seem a good test case for such a comparison.

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In this paper we determine the stacking fault energies by means of an *ab initio* method. Section 2 explains the structure of the different stacking faults of the FCC structure. Section 3 gives an account of the calculational procedure used, including some convergence tests. In section 4 the stacking fault energies are determined directly by performing calculations on large supercells and the results are compared to other recent theoretical studies [1, 2, 3, 4, 5]. It is shown that the calculated stacking fault energies are consistent with an Ising model with two interaction parameters. Finally the inter-planar relaxations are examined.

2. Stacking fault energies

In a close-packed stacking sequence of close-packed layers ($\langle 111 \rangle$ layers in the FCC structure) each of the layers can be positioned in three different positions usually referred to as A, B, and C. The unfaulted FCC structure corresponds to consecutively stacking in the ...ABCABC... stacking sequence, while the stacking sequence ...ABAB... gives the hexagonal close-packed structure. The two structures are illustrated schematically in figure 1. Apart from these two structures, three planar defects are of interest. (i) The *twin stacking fault*, ...ABCACBA..., where the FCC stacking sequence is reversed at one $\langle 111 \rangle$ plane. This is the ideal $\Sigma 3$ tilt grain boundary or the 180° twist grain boundary. (ii) The *intrinsic stacking fault*, ...ABCACABC..., where one $\langle 111 \rangle$ plane is missing in the FCC stacking sequence. This is the defect, that appears between the two partials of a split edge-dislocation which has a $\langle 111 \rangle$ plane as its slip plane. (iii) The *extrinsic stacking fault*, ...ABCBABC..., where one extra $\langle 111 \rangle$ plane has been inserted in the FCC stacking sequence. The three stacking faults are illustrated in figure 1.

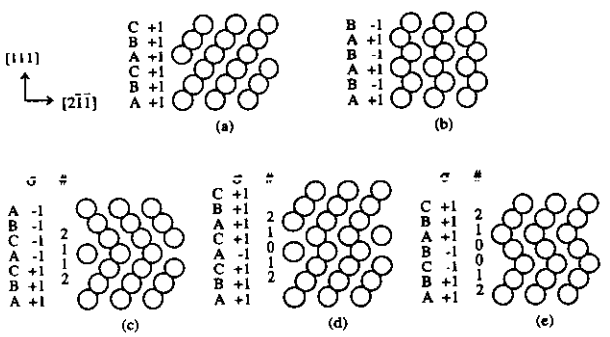

Figure 1. Schematic representations of (a) the unfaulted FCC structure, (b) the HCP structure, (c) the twin stacking fault, (d) the intrinsic stacking fault and (e) the extrinsic stacking fault The spins, σ, defined in section 4 and the layer separation numbers, #, used in table 2 are indicated.

3. The calculational method

We use the total energy pseudopotential method [6] with the Ceperley–Alder local exchange–correlation potential [7]. An *ab initio* non-local pseudopotential of the Kerker type [8] is used in the Kleinman–Bylander form [9]. The potential gives an equilibrium lattice constant of 3.95 Å in good agreement with the experimental value of 4.05 Å. The one-electron Schrödinger equations are solved in a plane-wave basis by means of the minimization technique based on conjugate gradients of Teter, Payne

and Allan [10]. Occupation numbers for the solutions are found by the Gaussian-smearing method of Fu and Ho [11] as modified by Needs *et al* [12]. The smearing width is 200 meV.

Plane waves with a kinetic energy up to 150 eV are included in the basis set. Convergence tests show that this is sufficient to determine energy differences to about 2 meV per atom for supercells relevant for the study of stacking faults.

The choice of k-points for integrating over the Brillouin zone is important for the convergence in total energy differences between different supercells. The optimal error cancellation is obtained when exactly the same k-points in absolute coordinates are used for both calculations. The supercells used for the stacking faults all have the same projection onto the layers but with a variable number of layers included in the cell. The Monkhorst–Pack grids [13] we use have 8×8 k-points in the planes. Due to the different numbers of layers in the supercells it is not possible to maintain a complete match of k-points in the direction of the stacking and a larger k-point density is therefore necessary in that direction. The number of k-points in the direction of the stacking is varied with the height of the supercell to keep an approximately constant k-point density. The density of k-points in this direction ends up being approximately 6 times higher than in the planes in order to ensure almost complete convergence in the k-space integration perpendicular to the planes. With this construction energy differences between different supercells can be determined to within a few meV.

The forces are calculated according to the Hellman–Feynman theorem when performing ionic relaxations. Forces and stresses calculated with non-converged wave functions serve to determine equilibrium positions and volumes while simultaneously finding the solution of the Kohn–Sham equations in the best Car–Parrinello [14] spirit.

4. Results and discussion

The structural energy difference between the FCC and HCP crystal structures can be calculated directly as the total energy difference between two supercells—one for the FCC and one for the HCP structure. With interlayer relaxations included, we find an energy difference between the two structures of 37 meV/atom = 86 mJ m^{-2}/atomic layer (the energy of the FCC structure being the lower). The relaxation effects are very small. If the planar and inter-planar distances are kept fixed at the equilibrium FCC values the energy difference is increased by only 0.8 meV/atom i.e. by a few percent.

We determine the stacking fault energies using supercells and therefore have to pay attention to exclude fault–fault interactions through the periodic boundaries. We have found a direct extrapolation of the results from larger and larger supercells the most appealing approach, as it is free from any assumptions of the nature of the interaction between the faults other than that it falls off smoothly for large fault–fault separations.

Figure 2 and 3 present the total energy per atom of calculations involving the three types of faults. The total energies are plotted against N_{atom}^{-1}, where N_{atom} is the number of atoms (and layers) per supercell. For $N_{\text{atom}}^{-1} = 0$ the total energy per atom, ξ_{FCC}, is that of an unfaulted FCC supercell geometry. The total energy of a super cell can be equated with a term summing ξ_{FCC} over all atoms plus a term summing the fault energy over all faults plus a term describing the total fault–fault interaction

energy. The energy per atom in the limit where the stacking faults do not interact can thus be written as:

$$E/N_{\text{atom}} = \xi_{\text{FCC}} + N_{\text{fault}}\Delta E_{\text{fault}}/N_{\text{atom}}, \tag{1}$$

where N_{fault} is the number of faults per supercell and ΔE_{fault} is the fault energy. For geometrical reasons $N_{\text{fault}} = 2$ in the calculations involving the twin fault and $N_{\text{fault}} = 1$ for the other two faults.

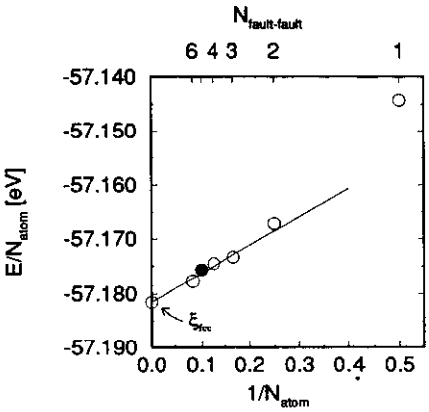

Figure 2. The (relaxed) total energies per atom for supercells containing two twin stacking faults plotted against N_{atom}^{-1}. The slope of the linear extrapolation through ξ_{FCC} for infinitely large supercells ($N_{\text{atom}}^{-1} = 0$) gives the twin stacking fault energy. Open circles indicate calculations for which the k-point separation perpendicular to the close-packed planes is 0.0574 Å^{-1}, the solid circle indicates a 0.0459 Å^{-1} separation. $N_{\text{fault-fault}}$ is the number of layers separating two faults.

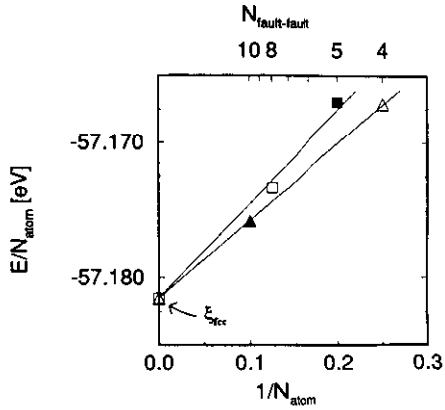

Figure 3. Same as figure 2, but for supercells containing one intrinsic stacking fault (squares) or one extrinsic stacking fault (triangles).

Figure 2 shows the results for the twin stacking fault for various unit cell sizes. The points in the figure all fall close to a straight line, equation (1), through $(N_{\text{atom}}^{-1}, E/N_{\text{atom}}) = (0, \xi_{\text{FCC}})$ for $N_{\text{atom}}^{-1} < 0.25$ i.e. for separations of the stacking faults of more than two layers. The deviations away from the straight line for large separations between the faults is of the size expected due to the finite k-point sampling. The energy points indicated with open circles in figure 2 are all calculated for a k-point separation of 0.0574 Å^{-1} perpendicular to the close-packed planes and thus benefit from optimal k-point error cancellation. The point indicated by a solid circle in the figure has been calculated with a k-point separation of 0.0459 Å^{-1}. The energies of the perfect FCC structure ($N_{\text{atom}}^{-1} = 0$) calculated with these two different k-point densities differ by only 0.3 meV/atom indicating a very good convergence of the k-space integration perpendicular to the planes. It is also seen that the solid circle lies close to the line defined by the open circles in the figure. Figure 3 shows similar results for intrinsic stacking faults (shown as squares in figure 3) and for extrinsic stacking faults (shown as triangles in the figure).

By fitting equation (1) to the points for small values of N_{atom}^{-1}, the energies of the three stacking faults in the limit of infinite large supercells can be evaluated to 27 meV/atom, 69 meV/atom and 61 meV/atom for the twin, intrinsic, and extrinsic stacking faults respectively (the unit 'meV/atom' denotes *'meV per atomic area'*, $A = \sqrt{3}a_0^2/4 = 7.1\,\text{Å}^2$, of the interface). The uncertainty in these numbers as estimated from the extrapolation procedure is around 10%. The effects of interlayer relaxations on the stacking fault energies are very small. As discussed for the HCP–FCC structural energy difference the changes in the energies are only of the order of a few percent and are therefore unimportant in a discussion of the energetics. However, the relaxations follow a particular pattern which gives some information about the interatomic interactions near the stacking faults which we shall discuss later.

Table 1 shows the calculated values of the stacking fault energies compared with other recent theoretical estimates and experimental values. Our results are seen to be in reasonable agreement with the experimental values. MacLaren *et al* [1] used the LKKR method to calculate the twin fault energy [15]. This was calculated as the difference between self-consistent calculations for two different structures. Later Crampin *et al* [2], still using the LKKR method, took advantage of the force theorem to avoid iterating to self-consistency and performed a better Brillouin zone sampling. The LKKR studies do not involve supercells and thus give energies for isolated stacking faults which do not require extrapolation. Xu *et al* [3] performed all-electron LMTO calculations in the atomic-sphere approximation. They use fault–fault separations of up to 7 layers, which according to our studies should suffice for avoiding dominant fault–fault effects. Denteneer and Soler [4] used the APW method together with an assumed fault–fault interaction form to determine the interaction terms and from these they obtained estimates for the isolated fault energies. Their APW calculations only include fault–fault separations up to two layers. Wright *et al* [5] in their pseudopotential study used supercells with a separation between the stacking faults up to 11 layers. They estimated the energies of isolated stacking faults by averaging over the energies of differently separated faults. As can be seen from the table all of the calculations except the one by Xu *et al* [3] agree within what can be expected from the numerical uncertainty due to the finite basis sets and convergence in the Brillouin zone integration. There seems to be no simple explanation why the results from the LMTO calculation [3] do not agree with the others since the basic asumptions of linearization in energy and the use of the atomic-sphere approximation are common to some of the other calculations.

Table 1 also includes the results of a set of calculations done with the local, empirical pseudopotential due to Heine and Abarenkov [16]. This potential has been used in recent studies [17, 18] and gives reasonable values for the equilibrium lattice constant, the bulk modulus, the cohesive energy and some frozen phonon frequencies (see [17]; this is also verified in our studies). However, as can be seen from the table the stacking fault energies calculated with this potential are about a factor two too small compared with the *ab initio* results. The former properties are governed by nearest-neighbour effects. The latter properties, for which the potential fails, are a result of subtle third-nearest-neighbour changes in atomic environment. We are therefore led to conclude that the empirical potential is not well suited for studies of such delicate long-range effects. For a review of stacking fault energies determined by use of empirical pseudopotentials—see Simon [19].

The quality of the linear fit, equation (1), with the energy points of small N_{atom}^{-1}

Table 1. Calculated results for the stacking fault energies and the FCC–HCP structural energy difference compared with other recent *ab initio* calculations and values derived from experiments [20]. Also included are the results obtained using the empirical potential due to Heine and Abarenkov [16].

	HCP ($mJ\,m^{-2}$/layer)	Twin ($mJ\,m^{-2}$)	Intrinsic ($mJ\,m^{-2}$)	Extrinsic ($mJ\,m^{-2}$)
This work	86	60	156	138
MacLaren *et al* [1]	—	59	—	—
Crampin *et al* [2]	—	56	124	118
Xu *et al* [3]	178	130	280	260
Denteneer and Soler [4]	71	54	126	108
Wright *et al* [5]	86	74	161	151
Empirical potential	50	34	66	55
Experimental [20]	—	75	166	—

in figure 2, indicates a rather short-ranged fault–fault interaction, and it is therefore natural to see to what degree a few parameter model can describe these interactions.

We follow the technique used by Cheng *et al* [21] in their SiC studies and which was also used for the determination of stacking fault energies by Denteneer *et al* [4]. The spin, σ_i, of the i'th layer is defined as $+1$ or -1 depending on the position of the $i+1$th layer. σ_i is $+1$ if the ith and $i+1$th layers follow the ABC stacking sequence and -1 if they follow the CBA stacking sequence—see figure 1. The total energy per atom is then expanded in pairwise interactions between the layers as

$$\frac{E}{N_{\text{atom}}} = \xi_0 - \sum_{n=1}^{n_{\max}} \frac{1}{N_{\text{atom}}} \sum_{i=1}^{N_{atom}} J_n \sigma_i \sigma_{i+n} \tag{2}$$

where i denotes the ith layer and n runs from 1 to the maximal range of the interaction, $n_{\max}$.

If we consider only the $n=1$ and $n=2$ terms the stacking fault energies are:

$$\Delta E_{\text{twin}} = 2J_1 + 4J_2 \qquad \Delta E_{\text{intr}} = 4J_1 + 4J_2 \qquad \Delta E_{\text{extr}} = 4J_1 + 8J_2.$$

If one determines the two parameters, J_1 and J_2 from three elementary calculations of the supercells of ABC, AB and ABCB stacking (these already appear in figure 2 at $N_{\text{atom}}^{-1}=0.0$, 0.5 and 0.25), one gets: $J_1 = 18.65$ meV and $J_2 = -2.125$ meV. These values give 29 meV/atom, 66 meV/atom and 58 meV/atom for the twin, intrinsic and extrinsic stacking fault energies respectively. These numbers derived from supercells of only up to four layers height correspond well to the values obtained above when using larger cells and the extrapolation procedure. We therefore conclude that the effects of stacking faults are indeed screened out over a few layer distances and the approach used by Denteneer and Soler [4] where the stacking fault energies are extracted from calculations on rather small supercells is justified.

We now turn to a discussion of the inter-planar relaxations. In table 2 the relative changes in the inter-planar separations in % are given for the largest supercells of figure 2 and 3. The layer separations are defined in figure 1.

The relaxations of the inter-planar separations are generally seen to be very small i.e. less than one percent in all cases. A general trend that is seen is that the relaxation of the separation denoted by 1 is about a 0.7% expansion and of separation

Table 2. The relative layer separation changes in %. The separations are defined in figure 1.

Separation #	HCP	Twin	Intrinsic	Extrinsic
0	0.7	—	0.9	0.1
1	—	0.7	0.7	0.6
2	—	−0.4	−0.3	−0.3

2 about a 0.3% compression. The relaxation of separation 0 varies considerably from the intrinsic to the extrinsic stacking fault, but is an expansion in both cases. Altogether the relaxations are seen to be consistent with a picture of rather short-ranged interactions between the layers. According to the Ising model there is an energy cost of $2J_1$ (neglecting for the moment the smaller J_2) associated with a configuration where two layers of the same type (A, B, or C) are separated by one other atomic layer. The positive sign of J_1 therefore indicates that the effective interaction between one layer and another of the same type two layer separations away is repulsive. The relaxations show that a repulsive force between two such layers also exists leading to the expansions. The small contractions of the subsequent layer separations can at least partly be explained through a push–pull effect: If we for instance consider the twin stacking fault (figure 1) the C layers next to the central A layer move away from each other due to the repulsion. However, the subsequent B layers do not shift rigidly together with the C layers because of the interactions between the B layers and the central A layer. The distance between the C and B layers (separation 2 in table 2) will therefore decrease slightly. That the relaxations of the inter-planar separations can indeed be understood in terms of pairwise layer interactions is seen by the interesting point that determining the relaxations within a pair potential model gives the same signs for the relaxations and roughly the same magnitudes [22].

5. Conclusion

The stacking fault energies for aluminium have been determined using the total energy pseudopotential method. Supercell effects are controlled by extrapolating the values of finite size cells to infinite cell sizes. The observed short-ranged fault–fault interactions are consistent with an Ising model with only two effective interaction parameters. The relaxations exhibit a systematic pattern related to the effective interactions between the layers. A comparison with other recent theoretical estimates and experimental values for the stacking fault energies shows a general agreement between the different approaches.

Acknowledgments

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