



Aluminium interatomic potential from density functional theory calculations with improved stacking fault energy

To cite this article: Xiang-Yang Liu *et al* 2004 *Modelling Simul. Mater. Sci. Eng.* **12** 665

View the [article online](#) for updates and enhancements.

You may also like

- [Transcutaneous high-frequency alternating current for rapid reversible muscle force reduction below pain threshold](#)
Yushin Kim, Thomas C Bulea and Hyung-Soon Park
- [A ternary EAM interatomic potential for U–Mo alloys with xenon](#)
D E Smirnova, A Yu Kuksin, S V Starikov et al.
- [A KIM-compliant *potfit* for fitting sloppy interatomic potentials: application to the EDIP model for silicon](#)
Mingjian Wen, Junhao Li, Peter Broomer et al.

Aluminium interatomic potential from density functional theory calculations with improved stacking fault energy

Xiang-Yang Liu^{1,4}, Furio Ercolessi^{2,5} and James B Adams³

¹ Computational Nanoscience Group, Motorola, Inc., Los Alamos, NM 87545, USA

² Department of Physics, University of Udine, I-33100 Udine, Italy

³ Department of Chemical and Materials Engineering, Arizona State University, Tempe, AZ 85287-6006, USA

Received 3 February 2003

Published 20 May 2004

Online at stacks.iop.org/MSMSE/12/665

DOI: 10.1088/0965-0393/12/4/007

Abstract

A new Al potential with improved stacking fault energy is constructed using the force-matching method. The potential is fitted to an *ab initio* forces database and various experimental data. By using a slightly larger cut-off, we found that the new potential gives the relaxed stacking fault energy in the experimental range without changing the excellent thermal and surface properties of the original force-matching Al potential given by Ercolessi and Adams (1994 *Europhys. Lett.* **26** 583).

Atomistic computer simulations have rapidly progressed in recent years. Empirical potentials have become more reliable, due to the advent of new many-body potential forms like the embedded atom method (EAM) [1] or the glue model [2]. These potentials are generally reliable for modelling crystal thermodynamics, crystal defects, surfaces and even liquids. The major advantage of empirical potentials is their computational efficiency, which allows the treatment of large systems of thousands to millions of atoms, provided the range of the potentials is kept sufficiently short.

However, Al is known to be more difficult to model with EAM potentials than noble metals such as Cu and Ag. There are at least six previous sets of EAM Al potentials. Voter and Chen [3] developed an EAM potential for Al by fitting to the lattice parameter, cohesive energy, elastic constants, vacancy formation energy, diatomic bond energy and diatomic bond length. The fits to elastic constants were all within 6%. However, the stacking fault energy (which the potential was not fit to) is roughly half of the experimental value. Foiles and Daw [4] fitted potentials for Ni₃Al, which predicted the Ni₃Al elastic constants well but had a 60% error for C₄₄ of pure Al. Later, Oh and Johnson [5] developed a short-ranged EAM Al potential, which also had poor elastic constants; for example, the anisotropy ratio ($2C_{44}/(C_{11} - C_{12})$)

⁴ Present address: Focus Center-New York, Rensselaer: Interconnections for Gigascale Integration, Rensselaer Polytechnic Institute, Troy, New York 12180-3590, USA.

⁵ Also at: Unità INFN/SISSA, I-34014 Trieste, Italy.

was off by 100%. To improve these, Rohrer [6] developed a new set of Al potentials (for Al–Cu–Ag ternary alloy potentials) which fits elastic constants and the stacking fault energy accurately. Although the surface properties are not well modelled by this potential, it is very useful for modelling the bulk properties at low temperatures. Ercolessi and Adams [7] tested the thermal behaviour of the potentials (except the potential by Oh and Johnson) and found that they described the thermal expansion and the melting temperature poorly.

An Al potential was previously constructed by Ercolessi and Adams [8] (E–A) using the force-matching method. In that approach, the empirical and *ab initio* methods are coupled very closely so as to use the advantages of both. The forces database includes samples from many different structures at finite temperatures, including bulk, cluster, liquid and surface structures. For each of these structures, *ab initio* molecular dynamics runs were performed in order to obtain a number of different samples. Extensive sampling on a variety of different geometries is crucial to achieve accurate modelling and high transferability. The resulting E–A potential was found to be highly reliable for many properties.

A study of Al dislocation core structures by Mills *et al* [9] found that the core structure of the Lomer dislocation predicted by the E–A potential is in good agreement with high-resolution transmission electron microscopy (HRTEM) observations. However, for a 60° dislocation the E–A potential predicted a somewhat wider core structure than experiment. This is due to the lower stacking fault energy (SFE) predicted by the E–A potential. Accurately predicting the correct SFE is desirable in modelling studies of extended defects, such as dislocations and grain boundaries. In this paper, we construct a new modified aluminium potential with a higher SFE energy. We use this potential to further develop an Al–Mg alloy potential for grain boundary simulations; we had briefly mentioned the Al potential with improved SFE energy in a previous publication [10]. In this short paper, the fitting detail and results of the new Al potential is described. This new potential should also be useful in other simulation studies, e.g. thin film depositions, dislocations in Al, etc. We also noticed that Mishin *et al* [11] also recently developed an Al potential specifically fitted to a higher SFE, which parallels our work. The potential was fitted to experimental data and a set of an *ab initio* structural energies instead of an *ab initio* forces database.

The detailed methodology of the force-matching method can be found in [8]. For the reader's convenience, we briefly overview the method here and concentrate on the details specific to fitting Al potentials. Potentials are generally defined by a number of single variable functions. For the EAM potential

$$E_{\text{tot}} = \sum_i F_i(n_i) + \frac{1}{2} \sum_{i \neq j} V_{ij}(R_{ij}) \quad (1)$$

and

$$n_i = \sum_{j \neq i} \rho_j(R_{ij}) \quad (2)$$

is defined by a pair potential $V_{ij}(R_{ij})$, an embedding energy $F_i(n_i)$, and an electron density function $\rho_j(R_{ij})$. Each function is described as a set of points that are connected by cubic splines. Let α indicate the entire set of L parameters $\alpha_1, \dots, \alpha_L$, which are used to characterize the functions. To determine the optimal set $\langle \alpha \rangle$, we try to match the *ab initio* forces for a large set of different configurations and simultaneously fit to various experimental data. Mathematically, this reduces to a non-linear minimization problem in an L -dimensional space with an objective function given by

$$Z(\langle \alpha \rangle) = Z_{\text{forces}}(\langle \alpha \rangle) + Z_{\text{experimental}}(\langle \alpha \rangle) \quad (3)$$

where Z is the mean square error in the fitting.

The forces data [12] are extracted by trajectories of molecular dynamics simulations using the local orbital density functional scheme [13]. The validity of the local orbital method is shown by a prediction of the lattice constant of 3.94 Å and bulk modulus of 0.812 Mbar for a confinement radius of 6.0 times the Bohr radius, compared with the experimental value of the lattice constant of 4.032 Å and bulk modulus of 0.809 Mbar. The configurations of the forces used in the Al potential fitting are the same as before [8]. These are bulk structures of 107 atoms with a vacancy at 100 K, 7 out of 35 MD steps; same bulk system at 1750 K (undergoing melting), 10 out of 100 MD steps; a (100) surface slab of 108 atoms at 100 K, 13 out of 39 MD steps; an equilibrated bulk liquid of 108 atoms at 2650 K, 20 out of 121 MD steps; a cluster of 150 atoms at 1000 K, 10 out of 100 MD steps; and the same cluster system in liquid state at 2200 K, 25 out of 465 steps. These involve a total of 85 configurations with 10 633 force vectors included. The forces database consisted of three position vector components and three corresponding force vector components for each atom at each of the 85 configurations. With a given parameter set (α) for describing the potential, the forces on the atoms in each configuration were calculated and compared to the *ab initio* forces during the fitting process. One open question regarding the force-matching method in constructing EAM potentials is the minimal dataset required of the first-principles force data. In our work, we conveniently designed the sampling space to include as much as possible the different geometries for accurate modelling and high transferability.

Besides the forces database, there are 32 other pieces of input data included in the fit. These are the cohesive energy, lattice constant, unrelaxed vacancy formation energy, unrelaxed (111) intrinsic stacking fault energy, unrelaxed (111) surface energy, elastic constants, the energy and pressure of the universal equation of state [14] at 11 different spacings ($a/a_0 = 0.90, 0.94, 0.97, 1.05, 1.11, 1.20, 1.30, 1.40, 1.50, 1.60, 1.75$), and the constraints related to the invariance of the potential.

Since the SFE is directly related to the energy difference between the hcp and the fcc crystal structures, increasing it means pushing up the energy of the hcp phase with respect to the fcc one. With this class of potentials, where no angular terms are present, these two crystal structures can be differentiated only in terms of the difference in the neighbour shells around a given atom. In fcc and hcp the first two shells are identical, so one needs at least third neighbour potentials to avoid degeneration. The original potential, with a cut-off at 5.56 Å, allowed for a difference, as fcc has 24 neighbours at 4.94 Å while hcp has 2 at 4.66 Å, 18 at 4.94 Å and 12 at 5.46 Å. However, all these distances are relatively close to the cut-off distance, where $\rho(r)$ is forced to go to zero smoothly. According to equation (2), the resulting difference in the total density n_i is modest, so that fitting a large SFE remains difficult. For this reason we have decided to extend the range to 6.06 Å, including an extra fcc and hcp shell at 5.70 Å (containing 12 and 6 atoms, respectively). But more importantly, the increased cut-off distance allows $\rho(r)$ to be larger in the important 4.6–5.5 Å range, and therefore the difference in n_i between the two structures can be made larger. Similar considerations hold for the pair potential part.

The original Al (E–A) potential was parameterized by a total of 40 parameters, 14 for $V(r)$, 12 for $F(n)$, and 12 for $\rho(r)$, with a cut-off value of 5.56 Å. In the modified Al potential, there are 43 parameters in total, 17 for $V(r)$, 12 for $F(n)$, and 14 for $\rho(r)$, with a cut-off value of 6.06 Å.

The spline parameters for the modified Al potentials are given in table 1, presented in tabular format. The Al properties predicted with the Al potential models and corresponding experimental values are presented in table 2. In the E–A potential, the rms deviation of the forces was 18.0%, which is 0.17 eV Å^{-1} of the average force of 0.92 eV Å^{-1} . The potential predicts remarkably good thermal properties (for details, see [8]) and surface properties. The

Table 1. Spline parameters for the modified AI potential functions. The spline function is calculated from: $s(x) = y(i) + b(i)(x - x(i)) + c(i)(x - x(i))^2 + d(i)(x - x(i))^3$ for $x(i) < x < x(i + 1)$, where $x(i)$ is the spline knot position, $y(i)$ is the function value at the knot, $b(i)$, $c(i)$ and $d(i)$ are the derivative coefficients that construct the spline function. (a) The spline parameters for pair potential $V(r)$, $x(i)$ in Å and $y(i)$ in eV. (b) The spline parameters for electron density $\rho(r)$, $x(i)$ in Å and $y(i)$ in arbitrary units. (c) The spline parameters for embedding energy $F(n)$, $x(i)$ in arbitrary units and $y(i)$ in eV.

$x(i)$	$y(i)$	$b(i)$	$c(i)$	$d(i)$
(a)				
2.0210	2.0051	-7.2241	9.3666	-4.3827
2.2730	0.7093	-3.3383	6.0533	-4.8865
2.4953	0.2127	-1.3713	2.7940	-2.3363
2.7177	0.0202	-0.4753	1.2357	-1.2893
2.9400	-0.0386	-0.1171	0.3757	-0.2907
3.1623	-0.0492	0.0069	0.1818	-0.3393
3.3847	-0.0424	0.0374	-0.0445	-0.0367
3.6070	-0.0367	0.0122	-0.0690	-0.2290
3.8293	-0.0399	-0.0524	-0.2217	0.4667
4.0517	-0.0574	-0.0818	0.0895	0.2227
4.2740	-0.0687	-0.0090	0.2381	-0.3170
4.4963	-0.0624	0.0499	0.0266	0.0796
4.7187	-0.0492	0.0735	0.0797	-0.2031
4.9410	-0.0311	0.0788	-0.0557	0.0980
5.1633	-0.0153	0.0686	0.0097	-0.2634
5.3857	-0.0024	0.0339	-0.1660	0.2612
5.6080	-0.0002	-0.0012	0.0083	-0.0102
6.0630	0.0000			
(b)				
2.0210	0.0824	0.0707	-0.1471	0.0554
2.2730	0.0918	0.0071	-0.1053	0.0460
2.5055	0.0883	-0.0344	-0.0732	0.0932
2.7380	0.0775	-0.0533	-0.0081	-0.0044
2.9705	0.0647	-0.0578	-0.0112	0.0432
3.2030	0.0512	-0.0560	0.0189	0.0040
3.4355	0.0392	-0.0465	0.0217	-0.0392
3.6680	0.0291	-0.0428	-0.0056	-0.0198
3.9005	0.0186	-0.0486	-0.0194	0.1593
4.1330	0.0082	-0.0318	0.0917	-0.1089
4.3655	0.0044	-0.0069	0.0157	-0.0242
4.5980	0.0034	-0.0035	-0.0012	0.0150
4.8305	0.0027	-0.0016	0.0093	-0.0218
5.0630	0.0025	-0.0008	-0.0059	0.0042
6.0630	0.0000			
(c)				
0.0000	0.0000	-18.4387	86.5178	-141.1819
0.1000	-1.1199	-5.3706	44.1632	-192.2166
0.2000	-1.4075	-2.3045	-13.5018	62.9570
0.3000	-1.7100	-3.1161	5.3853	-19.2831
0.4000	-1.9871	-2.6175	-0.3996	21.0288
0.5000	-2.2318	-2.0666	5.9090	-24.3978
0.6000	-2.4038	-1.6167	-1.4103	25.6930
0.7000	-2.5538	-1.1280	6.2976	-18.7304
0.8000	-2.6224	-0.4304	0.6785	1.6087
0.9000	-2.6570	-0.2464	1.1611	0.4704

Table 1. (Continued.)

$x(i)$	$y(i)$	$b(i)$	$c(i)$	$d(i)$
1.0000	-2.6696	-0.0001	1.3022	-2.3503
1.1000	-2.6589	0.1898	0.5971	-1.7862
1.2000	-2.6358	0.2557	0.0612	-1.7862

Table 2. Al properties predicted by EAM Al potential models compared to experimental data for the lattice constant, cohesive energy, bulk modulus, elastic constants, vacancy formation and migration energies, intrinsic (111) stacking fault energy, surface energy and surface relaxation between the two outermost layers for the (111), (100), and (110) surfaces, melting temperature and forces mismatch. All the energies are at 0 K and include relaxation effects.

	Experiment	New model	E-A [8]	V-C [3]	F-D [4]
a (Å)	4.032	4.032	4.032	4.05 ^a	4.05 ^a
E_c (eV/atom)	3.36	3.36	3.36	3.36	3.58 ^a
B (Mbar)	0.809 [15] ^b	0.809	0.809	0.79 ^a	0.81
C_{11} (Mbar)	1.180 [15] ^b	1.180	1.181	1.07	1.08
C_{12} (Mbar)	0.624 [15] ^b	0.623	0.623	0.652	0.68
C_{44} (Mbar)	0.325 [15] ^b	0.325	0.367	0.322	0.45
E_{vac}^f (eV)	0.66 [16]	0.68	0.69	0.63	0.62
E_{vac}^m (eV)	0.62 [17]	0.64	0.61	0.30	0.54
E_{sf} (mJ m ⁻²)	120–144 [18]	128	104	75	46
$\gamma(111)$ (meV Å ⁻²)	71–75 [19] ^c	57	54	51	31
$\gamma(100)$ (meV Å ⁻²)	71–75 [19]	63	59	53	34
$\gamma(110)$ (meV Å ⁻²)	71–75 [19]	69	65	60	37
$d_{12}(111)$ (%)	0.9 ± 0.7 [20]	0.3	0.9	-1.6	0.8
$d_{12}(100)$ (%)	-1.2 ± 1.2 [21]	-2.2	-1.5	-2.9	-1.7
$d_{12}(110)$ (%)	-8.5 ± 1.0 [22] ^d	-4.2	-4.6	-10.4	-5.4
T_m (K)	933.6	930	939	590	740
Error in forces		19%	18%	36%	45%

^a Fitted to a different set of experimental data.^b Extrapolated classically to $T = 0$ from data.^c This value includes the extrapolation to 0 K temperature.^d Reference [20] reports -8.4 ± 0.8 .

modified Al potential yields a higher relaxed SFE (128 mJ m⁻² compared to the experimental value of 120–144 mJ m⁻² [18]) by using a longer cut-off (6.06 Å instead of 5.56 Å) without changing the excellent thermal and surface properties of the original potential.

In conclusion, a new Al potential with improved stacking fault energy is constructed using the force-matching method. The potential is fitted to an *ab initio* forces database and various experimental data. By using a slightly larger cut-off, we found that the new potential gives the relaxed stacking fault energy in the experimental range without changing the excellent thermal and surface properties of the original Al potential developed by Ercolessi and Adams [8].

Acknowledgments

FE acknowledges support from MIUR COFIN. We thank Dave Drabold for providing the *ab initio* data. XYL thanks Hanchen Huang for helpful suggestions.

References

- [1] Daw M S and Baskes M I 1984 *Phys. Rev. B* **29** 6443
- [2] Ercolessi F, Tosatti E and Parrinello M 1986 *Phys. Rev. Lett.* **57** 719
- [3] Voter A F and Chen S P 1987 *Mater. Res. Soc. Symp. Proc.* **82** 175
- [4] Foiles S M and Daw M S 1986 *Phys. Rev. B* **33** 7983
- [5] Oh D J and Johnson R A 1988 *J. Mater. Res.* **3** 471
- [6] Rohrer C L 1994 *Modelling Simul. Mater. Sci. Eng.* **2** 119
- [7] Ercolessi F and Adams J B unpublished
- [8] Ercolessi F and Adams J B 1994 *Europhys. Lett.* **26** 583
- [9] Mills M J, Daw M S and Foiles S M 1994 *Ultramicroscopy* **56** 79
- [10] X-Y Liu and Adams J B 1998 *Acta Mater.* **46** 3467
- [11] Mishin Y, Farkas D, Mehl M J and Papaconstantopoulos D A 1999 *Phys. Rev. B* **59** 3393
- [12] Drabold D A private communication
- [13] Sankey O F and Niklewski D J 1989 *Phys. Rev. B* **40** 3979
- [14] Rose J H, Smith J R, Guinea F and Ferrante J 1984 *Phys. Rev. B* **29** 2963
- [15] Kamm G N and Alers G A 1964 *J. Appl. Phys.* **35** 327
- [16] Fluss M *et al* 1978 *Phys. Rev. B* **17** 3444
- [17] Schilling W 1978 *J. Nucl. Mater.* **69** 465
- [18] Westmacott K H and Peck R L 1971 *Phil. Mag.* **23** 611
Rautioaho R H 1982 *Phys. Status Solidi b* **112** 83
- [19] Tyson W R and Miller W A 1977 *Surf. Sci.* **62** 267
- [20] Adams D L, Nielsen H B and Andersen J N 1983 *Phys. Scr.* **T4** 22
- [21] Cheng H S *et al* 1990 *Nucl. Instrum. Methods Phys. Res. B* **45** 424
- [22] Noonan J R and Davis H L 1984 *Phys. Rev. B* **29** 4349