Modelling of heterogeneous clustering in aluminium

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Density functional calculations were performed to determine binding energies and the equilibrium lattice constant of aluminium for the conventional face centred cubic structure of 4 atoms. These were extended to supercells of respective size 2 x 2 x 2 and 3 x 3 x 3, that is, 32 and 108 atoms. A comparison was made between the total energy calculated for these structures and that obtained on the replacement of one of the aluminium atoms by a copper atom.

1.   Introduction

Precipitation hardening of alloys is carried out by the addition of small amounts of solute element to the pure metal. Improvements in electron microscopy techniques have allowed the imaging of clustering of the solute element. Fig.1 shows a High Resolution Transmission Electron Microscope (HRTEM) picture of such a particle of tin in an aluminium lattice.
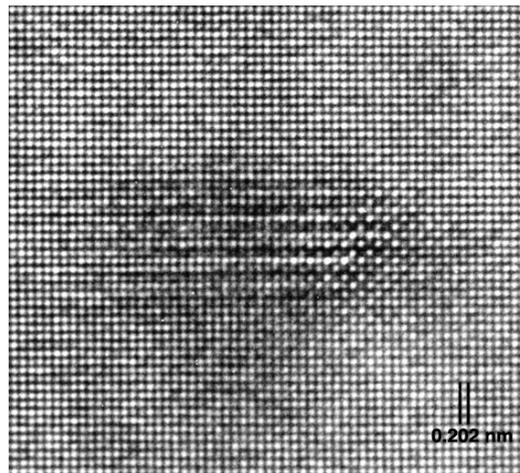

Fig. 1. HRTEM aluminium picture of a small platelet of the theta-prime phase (dark line contrast, left) attached to a particle of tin (bright white dots, right). [National Institute for Materials Science, Tsukuba, Japan field-emission gun 3000F JEOL].

With increasing computational power, atomic scale effects can now be better simulated to determine the nature of the hardening mechanism [1]. In the present work the plane wave pseudopotential density functional theory package fhi98md [2] has been used to study the beginning of clustering in aluminium. Initial convergence conditions have been tested by determination of binding energies for a variety of super cell sizes of the aluminium host crystal. These were compared with total energy calculations for single copper atom precipitates, which will lead to similar calculations for small size precipitates. Such local optimal determinations are seen as precursors to full Monte-Carlo calculations of the notional best local geometry for larger precipitates. Comparisons have also been made with results obtained from the Linear Augmented Plane Wave WIEN2K computer package [3] as plane

wave based pseudopotential methods such as the fhi98md package are known not to converge optimally for flat banded transition metals such as copper [4].

2.   Results and Discussions

Calculations were carried out for the conventional aluminium fcc cell of 4 atoms (Fig.2) and compared with those from larger supercells. For instance, Fig. 2 shows the 108 atoms of the 3 x 3 x 3 supercell containing 27 x 4 = 108 atoms. A solute atom would later replace one of these atoms.
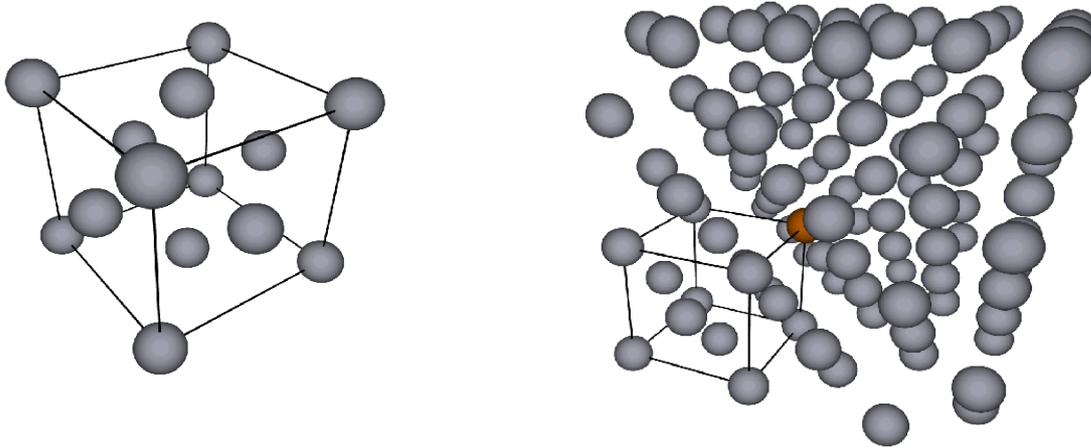

Fig. 2. Conventional fcc cell consisting of 4 atoms compared with supercell of length 3 x 3 x 3 and comprising 4 x 27 = 108 = 107 + 1 atoms.

By carrying out total energy calculations for different values of the lattice parameter it is possible to determine the equilibrium lattice constant. Fig. 3 shows the determination of the equilibrium lattice constant for the conventional aluminium lattice by a Murnaghan plot of the Total Energy as a function of the lattice parameter [5].
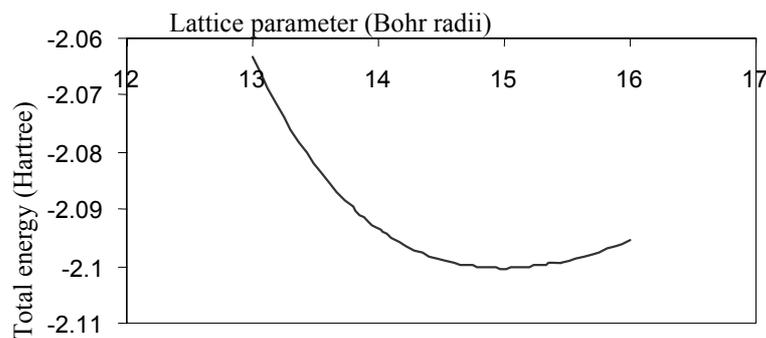

Fig. 3. Equilibrium lattice constant determination. Murnaghan plot of Total Energy (Hartree) as a function of lattice parameter (Bohr radii)

The Binding energy of the aluminium lattice could be determined by taking the difference between the Total Energy at the equilibrium lattice distance and at very large lattice distances as shown in Fig. 4.
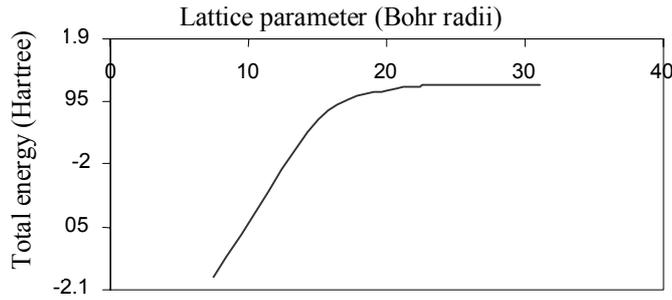
Fig. 4 Binding energy determination. Total Energy (Hartree) as a function of lattice parameter (Bohr radii).

Total Energy calculations were performed for the periodic object consisting of the supercell with one of the aluminium atoms replaced by a copper atom. Whilst the aluminium pseudopotential is part of the test data in the fhi98md package [2], it was necessary to calculate the copper pseudopotential separately [3]. Table 1 compares total energies and demonstrates that the introduction of the copper atom results in a greater binding energy. As the total energy has been deceased it is concluded that the addition of even a single copper atom has a hardening effect.

Atom number	E_{tot} (H)	E_{tot} (H)/ atom
4	-8.36	-2.09
32	-67.21	-2.09
108	-226.81	-2.09
31 + 1	-129.99	-4.06
107 + 1	-289.64	-2.69

Table 1. Comparison for total energy calculations of pure aluminium (first three rows) with copper atom replacement in respectively the 2 x 2 x 2 and 3 x 3 x 3 supercell.

In order to validate the copper calculation, comparisons were made with results from the WIEN 2K method. In particular convergence properties were checked (see Fig.5 which compares total energies for different values of the product of maximum wave number with the Muffin Tin Radius) together with the quality of the obtained electronic band structure (Fig. 6)
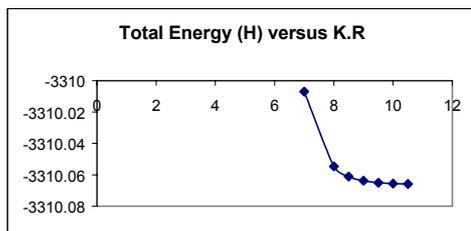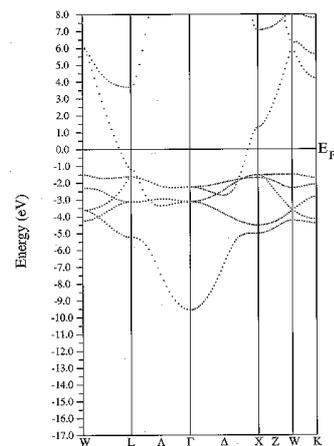

Fig. 5. WIEN 2K convergence calculation for Cu

Fig. 6 . Wien 2K Cu bandstructure

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