Computational Materials Science 85 (2014) 1–7

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Atomic ordering in liquid aluminium induced by substrates with misfits

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ARTICLE INFO

Article history:
Received 18 October 2013
Received in revised form 4 December 2013
Accepted 20 December 2013
Available online 11 January 2014

Keywords:
MD simulations
Interface
Lattice misfit
Atomic ordering

ABSTRACT

In this paper, the effect of the lattice misfit (*f*) between the crystalline substrate and solid Al on both atomic layering and in-plane atomic ordering of the Al atoms at the liquid Al/substrate interface with (1 1 1) orientation has been investigated using molecular dynamics (MD) simulations. It was found that the Al atoms became layered within 5–6 atomic layers at the interface, independent of *f*, however the in-plane ordering strongly depended on the magnitude of *f*. While $|f|$ is less than 3%, the pronounced in-plane ordering persists within a few atomic layers at the interface, where the Al atoms in the 1st layer are well localized at the equilibrium sites on the surface of the substrate. With a further increase in $|f|$, the in-plane ordering of the Al atoms decreases dramatically, and only a certain in-plane atomic ordering persists within the 1st atomic layer at the interface.

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1. Introduction

Knowledge of the atomic arrangement of liquid adjacent to a crystalline substrate is of importance for understanding heterogeneous nucleation, grain refinement, wetting, liquid phase joining and lubrication [1,2]. The atomic structure of liquid at interfaces has been a long-standing topic for theoretical analysis and experimental investigation [3–11]. Theoretical calculations have shown that the liquid will become layered at the free surface or at the interface with a solid wall, giving rise to an oscillatory density profile [3–7]. This phenomenon was attributed to the effect of the 'hard wall', against which the liquid could be packed in layers [12,13]. X-ray diffraction experiments [8–11,14–16] and atomistic simulations [17–26] demonstrated that the atoms of liquid metal became layered within a few atomic layers at the free surface or at the liquid/solid interface. However, the in-plane structure within these layers is still an open question [27]. Experimental observation [15] and atomistic simulation [28] have confirmed that the in-plane structure of liquid Ga at the free surface is similar to the bulk liquid without any obvious positional order, and Oh et al. [29,30] have provided evidence for the layering and in-plane ordering of liquid Al adjacent to α-Al_2O_3 based on direct high-resolution transmission electron microscopy (HRTEM) observation. In addition, high-energy X-ray reflection [31] and atomic simulation [32] have revealed that the layering at the liquid Hg/α–Al_2O_3 interface is less pronounced than that at the free surface of liquid Hg. It seems likely that the solid substrate has an important effect on the in-plane structure of a liquid metal at the interface.

Given the difficulties associated with the design of experiments that are able to probe the interface at an atomic length scale, most current knowledge on the subject has been derived from atomistic simulations [2]. Atomistic simulations offer direct access to microscopic details of the liquid/solid interface. Geyermans et al. [33] examined the interface between solid Cu and liquid Al with MD simulation via adapted *n*-body potentials, and revealed that the presence of a solid Cu wall induced significant layering in the density profiles of liquid Al at the interface, which was largely independent of the surface orientation of the substrate. Hashibon et al. [2,34,35] used an embedded atom method (EAM) to study a model liquid metal/substrate interface. They assumed that the substrate was composed of static atoms positioned in an ideal lattice with either a face-centred cubic (fcc) or body-centred cubic (bcc) symmetry but with different terminating planes, and that the liquid of the same element was introduced adjacent to the solid. This study not only confirmed an exponential decay of the density profile of liquid Al at the interface, regardless of the orientation of the substrate, but also found that there was a far greater in-plane ordering of the liquid in contact with a bcc (100) substrate than a bcc (1 1 0) substrate. Pablo et al. [36] examined the interface between a solid Cu (1 1 1) plane and liquid Pb with an EAM potential, and observed a "prefreezing" layer of crystalline Pb within approximately 2–3 atomic layers at the interface. These findings indicated that substrate orientation had a strong influence on the degree of the in-plane atomic ordering, but not on the atomic layering.

Interfacial atomic ordering of the liquid could have a significant influence on the nucleation behaviour of alloy melts [37]. The hypernucleation hypothesis [38,39] suggested that a layer of quasi-solid formed on the surface of TiB_2 particles in Al–Ti–B inoculated Al melts even above their melting temperature, and the

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http://dx.doi.org/10.1016/j.commatsci.2013.12.042

adsorption model [40,41] suggested that a layer of a new crystalline phase can be adsorbed at the surfaces of the substrates beyond a critical undercooling. The quasi-solid layer could serve as a template for the growth of a new phase at a very small undercooling [42,43]. The recent MD simulations [25,26] indicated that the Al_3Ti $(1\,1\,2)$ substrate effectively initiated the free growth of α-Al phase in the melt but neither the Al_3Ti $(1\,1\,0)$ nor the Al_3Ti $(1\,0\,0)$ substrate, where the lattice misfit of $Al(1\,1\,1)//Al_3Ti(1\,1\,2)$ was substantially smaller than that of $Al(1\,1\,0)//Al_3Ti(1\,1\,0)$ or $Al(1\,0\,0)//Al_3Ti(1\,1\,0)$. For a very potent nucleating particle, the lattice misfit is typically less than 10% [44]. So far, our knowledge of the effect of physical and chemical properties of the substrate on the structure of liquid metals at the interface is very limited.

The objective of the present work was to systematically investigate the effect of lattice misfit on the layering and the in-plane ordering of the Al atoms at the liquid Al/substrate interface, using the MD simulations for a model system of a liquid Al/fcc substrate with varied f.

2. Simulation method

The MD simulation was conducted for the system of liquid Al and a substrate with an fcc structure and a $[1\,1\,1]$ surface orientation. Fig. 1 is an illustration of the simulation cells, where the dark balls represent fcc substrate atoms and the light balls represent the Al atoms. We used the EAM potential for Al developed by Zope and Mishin [45] to model the interatomic interactions. The predicted melting temperature for pure Al is 870 ± 4 K with their EAM potential [25]. The initial configuration of the Al sample had a lattice parameter $a = 4.12$ Å, which corresponded to the value obtained at the calculated melting point. The material of the substrate was Al, but with a specific negative f $(0 \sim -8\%)$. The lattice misfit is defined as:

$$f = \frac{d_m - d_{sb}}{d_m}, \tag{1}$$

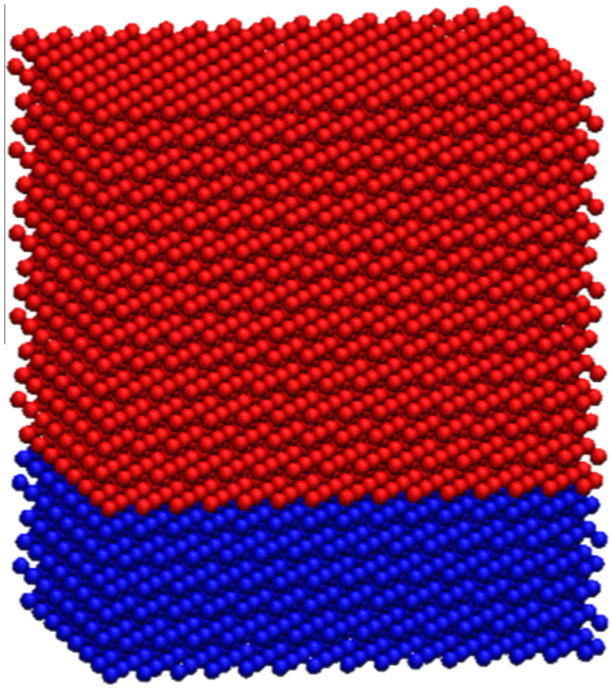

Fig. 1. An illustration of the simulation cell. The dark balls represent fcc substrate atoms, and the light balls represent the Al atoms. The size of the cell is 48 $[1\,1\,\bar{2}] \times 30$ $[\bar{1}\,1\,0] \times 21$ $[1\,1\,1]$ (d is the inter-planar spacing of the corresponding planes).

where d_m and d_{sb} are the corresponding interatomic spacings along the close packed directions on the specified planes of the solid metal (the equilibrium solid phase solidified from the liquid) and the substrate, respectively. The terminating plane of the substrate was $(1\,1\,1)$, with the z axis normal to the $(1\,1\,1)$ plane. The dimensions of the substrate were 48 $[1\,1\,\bar{2}] \times 30$ $[\bar{1}\,1\,0] \times 6[1\,1\,1]$, and the initial dimensions of the Al sample were 48 $[1\,1\,\bar{2}] \times 30[\bar{1}\,1\,0] \times 15[1\,1\,1]$. Periodic boundary conditions were imposed in the $x([1\,1\,\bar{2}])$- and $y([\bar{1}\,1\,0])$-dimensions. The z-dimension ($[1\,1\,1]$) was considered as a dynamic variable, and above the Al sample a vacuum region was inserted with periodic boundary conditions in all directions. The extent of the vacuum region in the z-direction was 25 times that of the substrate plus the Al sample. During the simulation the Al atoms were allowed to move freely under the effect of the interatomic potential, and the substrate atoms were frozen to a perfect fcc lattice configuration with a specified lattice parameter. The substrate atoms were excluded from the equations of motion, but the forces they exerted on the adjacent atoms were included. In this way, the substrate could be considered as a dissimilar material (e.g., ceramic) with a much higher melting temperature.

The MD simulations were performed using the DL_POLY_3.10 MD package [46]. The equations of motion were integrated by means of the Verlet algorithm with a time step of 0.001 ps, and the Berendsen NVT ensemble was used for the temperature control. The sample was equilibrated first at 0 K for 100,000 MD steps, and then heated to a temperature of 1400 K by steps of 50 K, each lasting 100,000 MD steps. For the system with $f = 0\%$, the melting point of the Al sample has been determined to between 850 K and 900 K with the coexistence approach, which agreed qualitatively with the calculated melting point (870 K) with this EAM potential. The equilibrated configuration of the liquid Al was cooled to the desired temperature in steps of 50 K, and at each temperature step the system was allowed to run for 100,000 MD steps to equilibrate. During the equilibration stage, the total energy was monitored to verify the approach to equilibrium.

3. Results

3.1. Effect of lattice misfit on the atomic layering

The MD results [47] indicate that there is a potential energy minimum for a liquid atom at the surface of a crystal, which is provided by the location of the atoms at the surface of the crystal. An atom in the liquid at the interface can fall into this potential energy minimum with thermal velocity, and its behaviour may deviate dramatically from that in the bulk liquid.

Fig. 2 shows an example of the simulation cell with $f = 0\%$ equilibrated at a temperature $T = 1000$ K for 100,000 MD steps. The dark balls represent the substrate atoms, which are fixed at their initial positions during the simulations, while the light balls represent the Al atoms. A layered structure is observed for the Al atoms within a few atomic layers at the interface, and further away the atoms adopt the characteristics of the bulk liquid.

To characterize the atomic layering of the Al atoms at the interface, we calculated the z-dependent density profile. The density profile is an average number density of the atoms in a slice of width Δz centred around z, in which the simulation cell is divided into equal bins parallel to the rigid interface. The density profile is expressed as [34]:

$$\rho(z) = \frac{\langle N_z \rangle}{L_x L_y \Delta z}, \tag{2}$$

where N_z is the number of atoms between $z - \Delta z/2$ and $z + \Delta z/2$ at time t, and the angled brackets indicate a time average, and L_x and L_y are the x and y dimensions of the cell.

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