



Atomic ordering in the liquid adjacent to an atomically rough solid surface

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ABSTRACT

In this work, atomic ordering in the liquid adjacent to both crystalline and amorphous substrates with atomic level surface roughness was investigated systematically using molecular dynamics (MD) simulations. We found for the first time that increasing surface roughness of a crystalline substrate reduces both atomic layering and in-plane atomic ordering in the metallic liquid adjacent to the liquid/substrate interface. In addition, our MD simulation results revealed that the rough surface of an amorphous substrate eliminates completely in-plane ordering in the liquid regardless of surface roughness and reduces/eliminates atomic layering in the liquid depending on the level of surface roughness. This reduced atomic ordering in the liquid adjacent to an atomically rough surface can be attributed to the increase in mobility of atoms in the liquid compared with the case with a smooth crystalline surface. From the point of view of heterogeneous nucleation, in addition to the effect of lattice misfit investigated in our previous studies, this work provides further confirmation of the importance of structural templating as a mechanism for both prenucleation and heterogeneous nucleation. Furthermore, this work offers a new approach to impede heterogeneous nucleation by roughening the substrate surface at the atomic level.

1. Introduction

Atomic ordering in the liquid adjacent to a solid substrate has recently attracted increasing interest in the solidification research community, due to its implications for heterogeneous nucleation [1,2]. Such atomic ordering at temperatures above the liquidus has been referred to as prenucleation [3], which can be taken as a precursor for the subsequent heterogeneous nucleation process. The Epitaxial Nucleation model [4] suggests that heterogeneous nucleation proceeds through layer-by-layer growth by a structural templating mechanism. The crystal lattice in the substrate surface provides low energy positions where the adjacent liquid atoms can form a locally ordered structure, which in turn templates the formation of an ordered structure in the next layer and so on. The undercooling required for epitaxial nucleation is closely related to the compatibility of the crystal structures of the substrate and the solidified phase, which can be quantified by their lattice misfit. Therefore, one would expect that pronounced atomic ordering in the liquid at the interface, above the liquidus, can have a significant influence on the heterogeneous nucleation process. If atomic ordering at the interface is compatible with the crystal structure of the solid it would enhance heterogeneous nucleation by reducing the nucleation barrier; otherwise, incompatible atomic ordering at the interface would impede heterogeneous nucleation. Therefore, it is important, both scientifically and technologically, to have a good

understanding of how the chemical and/or physical properties of the substrate affect atomic ordering in the liquid at the interface and its implications for the heterogeneous nucleation process.

Both experimental observations [5–10] and atomistic simulations [11–16] have been conducted to understand atomic ordering in the liquid at liquid/substrate interfaces. These studies suggest that at temperatures above the liquidus the liquid atoms become layered within one or two nanometres of the interface (atomic layering) and that the atoms in individual atomic layer may have a substantially ordered structure (in-plane atomic ordering). Oh et al. [9,10] have provided firm evidence for atomic layering and in-plane ordering in liquid Al adjacent to α -Al₂O₃ substrates with a [0 0 0 1] surface orientation, through *in situ* observation by high-resolution transmission electron microscopy (HRTEM). In their MD simulations with adapted n-body potentials, Geyermans et al. [11] revealed that a solid Cu wall induces significant layering in the liquid Al at the interface, largely independent of surface orientation of the substrates. Using a semi-empirical potential of an embedded-atom method (EAM), Hashibon et al. [12,13] revealed an exponential decay of density profile in the liquid Al at the interface, and found that there is far greater in-plane ordering in the liquid in contact with a bcc (1 0 0) substrate than that in contrast with a bcc (1 1 0) substrate. These atomistic simulations offer access to microscopic details of atomic ordering in the liquid adjacent to the liquid/solid interface.

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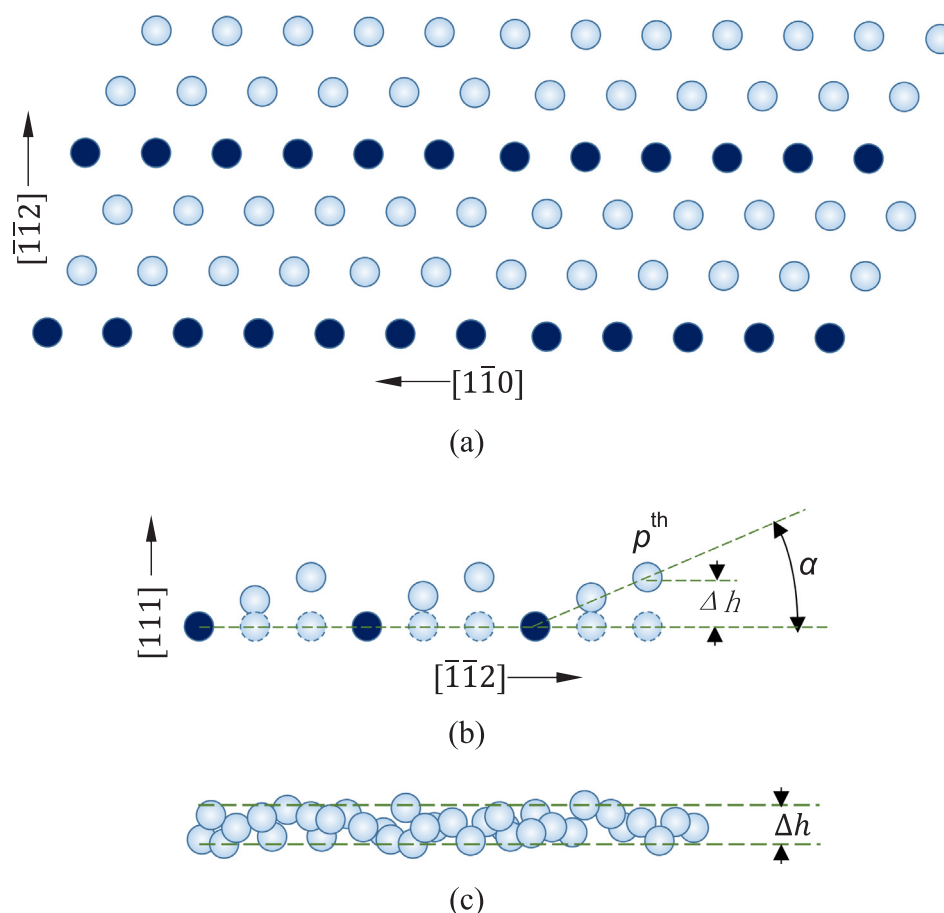


Fig. 1. Schematic illustration of the artificial construction of substrates with varied atomic level surface roughness. An artificially constructed crystalline rough surface with (111) surface orientation of an fcc Al substrate is viewed (a) from the [111] direction and (b) from the $[1\bar{1}0]$ direction, and (c) an artificially constructed amorphous Al rough surface is viewed from a direction perpendicular to its surface normal. Dark spheres represent atoms that remain in the original surface plane, and light spheres represent atoms that are displaced in a direction parallel to the surface normal to create surface roughness.

Table 1

Comparison of the physical properties of Al from the simulation in this study with that from experiments in the literature [25–27].

	This study	Experiments	Error (%)
Lattice parameter (Å)	4.0830	4.05 [25]	0.83
Melting point (K)	916.0 ± 13.5	933.45 [26]	–
B_0 (GPa)	79.07	79 [27]	0.013
C11 (GPa)	113.71 ± 1.04	114 [27]	–0.8
C12 (GPa)	61.69 ± 1.39	61.9 [27]	0.7
C44 (GPa)	31.16 ± 0.89	31.6 [27]	–5.2

The atomic ordering in a given liquid at the interface can be manipulated by changing the structure and/or chemistry of the substrate. The layering has been attributed to the ‘hard wall’ effect of the substrate surface [17], and theoretical calculations [18] suggest that the liquid has an oscillatory density profile at the interface with a structureless solid wall. The degree of the layering is usually independent of crystal structure [12], surface orientation [11,12] of a substrate with a smooth surface, and lattice misfit between the substrate and the solid phase corresponding to the liquid [14]. Atomic layering has even been observed in metallic liquids adjacent to their own surfaces by X-ray reflectivity measurements [17,19–21], and at the interface with the smooth surface of an amorphous substrate using the MD simulation [11]. All these studies suggest that the layering at the interface hardly changes by changing the substrates as long as the substrate surface is smooth. On the other hand, the in-plane atomic ordering at the interface has been attributed to the crystalline lattice in the surface of the

substrate, which provides potential low energy positions for the liquid atoms at the interface. Therefore, the in-plane atomic ordering is closely related to the crystal structure of the substrate [11–14]. Using MD simulations, it is found that the in-plane ordering persists within the first 3 atomic layers adjacent to an interface having a small lattice misfit, and becomes very weak, even in the 1st layer for substrates having a large lattice misfit [14]. This suggests that the in-plane atomic ordering can be manipulated by changing the crystallographic matching between the substrate and the solid upon solidification. In addition, we found recently that chemical interactions between the substrate and the liquid may further enhance or impede the structural effect on atomic ordering at the interface, including both layering and in-plane ordering [22].

The atomic ordering in the liquid at the liquid/substrate interface may be affected by the surface roughness of the substrate. To date, only a small number of studies on this topic have been reported in the literature. Using MD simulations, Geysers et al. [11] revealed that atomic layering is significantly weakened by increasing the surface roughness of an amorphous substrate, and even destroyed completely by the rough surface of a bulk amorphous substrate. Galea et al. [23] investigated the effect of atomic level roughness of crystalline substrates on slip length at the fluid/solid boundary during shear flow, by varying the size and spacing of substrate atoms at a constant packing fraction, and they found that the amplitude of the density oscillations at the interface increases with increasing smoothness of the surfaces. In both cases, however, the effect of surface roughness on the in-plane atomic ordering was not investigated. Therefore, it is desirable to clarify how atomic ordering (both layering and in-plane ordering) in the liquid at the interface is affected by the surface roughness of the

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