Effect of physical and chemical properties of the substrate on prenucleation

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Atomic ordering in the liquid adjacent to a solid substrate (i.e., prenucleation) has recently attracted increasing interest in the solidification research community, due to its implications for heterogeneous nucleation [1].

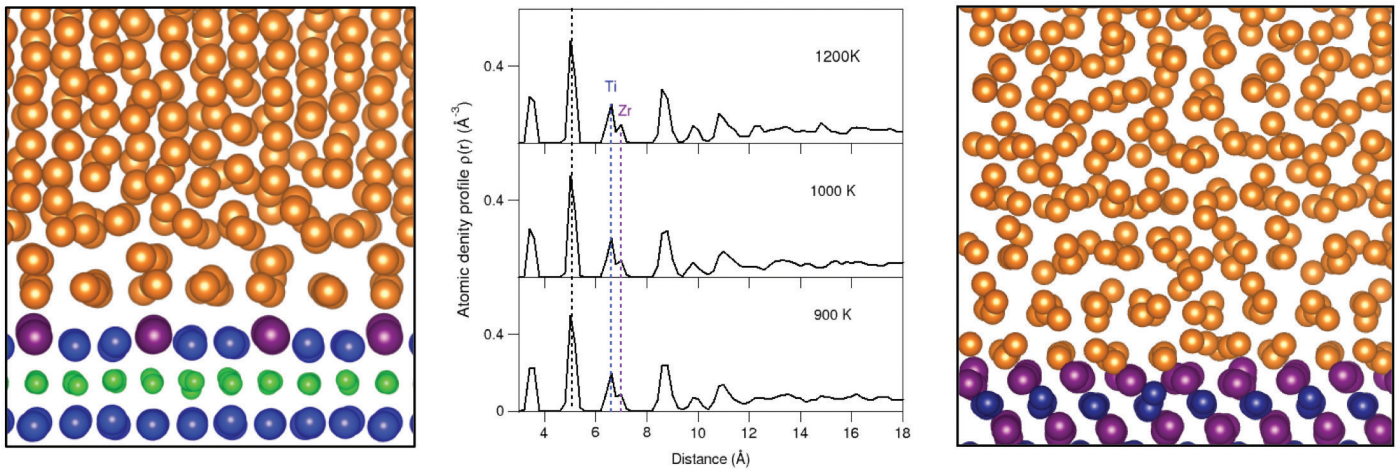

FIGURE 1. (a) Snapshot at 1000 K and (b) density profiles of liquid Al/TiB_2(0001) with $ZrTi_2$ 2DC (green, Ti; blue, Ti; purple, Zr), and (c) snapshot of liquid Mg/MgO(111) at 1000 K (dark blue, Mg; purple, O). In both cases the substrate surface becomes atomically rough.

It has been recognised that the atomic ordering is relevant to some properties of the substrate. However, full pictures about effects of its physical and chemical properties on the prenucleation, as well as its origin, are not yet established. In the past, we revealed that the layering at the interface is independent of the lattice misfit, and the in-plane atomic ordering degrades with an increase of the lattice misfit [2]. In this project, we intend to investigate the effect of physical (lattice parameter and surface roughness) and chemical natures of the substrate, and to clarify the interplay between structural and chemical effects of the substrate on atomic ordering. Classical and ab initio molecular dynamics (MD) simulations were carried out for the systems of liquid Al and substrates with varied lattice parameter, chemistry and/or atomic level surface roughness. Further, we analysed atomic ordering, mobility of the atoms, and charge transfer at the interface, using different techniques.

This project revealed the formation of a two-dimensional (2D) ordered structure in the liquid on the substrate surface through a structural templating mechanism, which is dependent on the lattice misfit. We also found that chemistry of the substrate produces noticeable effect on both layering and in-plane atomic ordering at the interface, where an attractive chemical interaction strengthens the atomic ordering, whilst a repulsive interaction weakens it. The enhanced atomic ordering for an attractive chemical interaction is attributed to reducing atomic mobility in the liquid due to a higher electron transfer across the interface. In addition, for the first time we demonstrate that increasing surface roughness of a crystalline substrate reduces both atomic layering and in-plane atomic ordering, and this can be attributed to the increase in atomic mobility. Furthermore, atomic level surface roughness was found in a monolayer of $ZrTi_2$ 2DC on TiB_2 substrate in liquid Al (Figures 1a and 1b) and at the liquid Mg/MgO{111} interface (Figure 1c). Such a rough substrate surface degrades dramatically the atomic ordering in the liquid at the interface, rendering the substrate impotent for heterogeneous nucleation.

This project provides insight on the effect of physical and chemical properties of the substrate on the prenucleation and subsequent heterogeneous nucleation. These are crucial to the development of more effective approaches to grain refinement during solidification through effective manipulation of the interplay between structural and chemical effects of the substrate on atomic ordering.

As part of the future research focus, we will examine the effect of atomic mobility at the interface on both prenucleation and heterogeneous nucleation in more practical systems, such as alloy melts containing native oxides.

REFERENCES:

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[2] H. Men and Z. Fan. Atomic ordering in liquid aluminium induced by substrates with misfits. *Computational Materials Science*, 85 (2014), 1. DOI: 10.1016/j.commatsci.2013.12.042.