A molecular dynamics study of structural templating during heterogeneous nucleation

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Heterogeneous nucleation theory is of fundamental importance in understanding solidification processes. However, the effect of physical and chemical properties of the nucleant particle on atomistic mechanisms of heterogeneous nucleation is so far poorly understood.

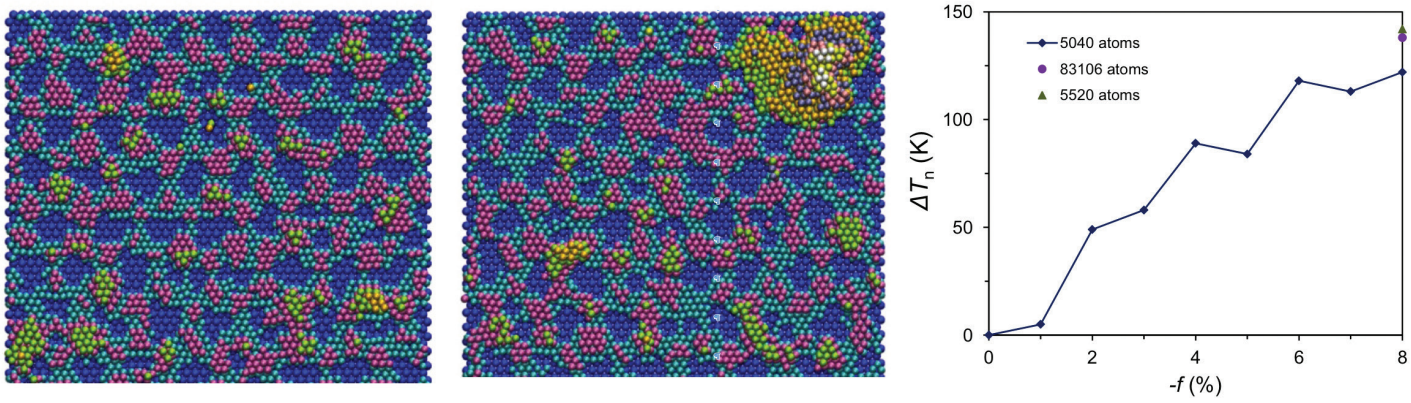

FIGURE 1. 2D ordered structures and new phase at (a) t = 200 ps and (b) 300 ps at the interface with a substrate having a lattice misfit f = -8 % at 744 K, and (c) undercooling for heterogeneous nucleation as a function of f. The heterogeneous nucleation proceeds by a structural templating mechanism.

In recent years, it has been realised that atomic ordering in the liquid adjacent to the liquid/substrate interface has significant implication on the heterogeneous nucleation processes [1]. In our previous work [2], we found that two-dimensional (2D) ordered structure formed at the interface even above the liquidus, i.e., prenucleation. The epitaxial nucleation model [3] suggests that prenucleation may provide a mechanism of structural templating for heterogeneous nucleation. In this study, we investigated the structural templating mechanism during heterogeneous nucleation using molecular dynamics (MD) simulation. The MD simulations were conducted for systems of liquid/substrate with varied lattice misfits, and EAM potentials were used to model the interatomic interactions.

In this study, we established an atomistic mechanism of heterogeneous nucleation through structural templating, i.e., a new phase was created by the formation of a 2D ordered structure at the interface. Solid atoms in ordered structure in the 1st layer continue the lattice of the surface layer of the substrate in either fcc or hcp stacking sequence, namely structure templating, at the prenucleation stage (Figure 1a). During the nucleation, ordered regions in the 2nd layer merge together to form the new phase by continuing the lattice of the first layer (Figure 1b). Simultaneously, shockley partial dislocations with predominant screw components were generated between the 1st and 2nd layers in the new phase, leading to a twist of a new phase relative to the substrate. Generation of the partial screw dislocations is largely responsible for nucleation barriers. Further, we found that density of dislocations increases with increasing lattice misfit, as well as undercooling of the nucleation (Figure 1c), i.e., the potency of the substrates degrades with an increase of lattice misfit. Thus, this study reveals the process of heterogeneous nucleation is closely relevant to the property of the substrates.

For the first time we revealed the process of heterogeneous nucleation at an atomic level through a structural templating mechanism, which would be generalised for other materials with varied structures and chemistry. This study has shed new light on the heterogeneous nucleation theory.

Thermodynamics and kinetics of heterogeneous nucleation need to be clarified for an in-depth understanding of the nucleation. Atomistic mechanism of heterogeneous nucleation in the systems of practical interests should be examined in the future to validate the structural templating mechanism.

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