Orientation dependent ordering of liquid Al atoms and oxygen transport mechanism at liquid Al/sapphire interfaces

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Understanding the nature of liquid/solid interfaces is important for many processes of technological interest, such as solidification, liquid phase epitaxy, wetting, liquid phase joining, lubrication and nanowire growth by vapour-liquid-solid (VLS) processes. We previously provided the first direct evidence for ordering of liquid atoms adjacent to an interface with a crystal, based on real-time HRTEM observations of aluminium/sapphire liquid/solid (LS) interfaces [1]. The present in-situ HRTEM study explores the orientation dependent ordering of liquid Al atoms on a sapphire crystal and reveals the oscillatory oxygen transport mechanism operating at the triple junction of vacuum/liquid Al/sapphire.

In-situ heating TEM experiments were performed in the MPI-Stuttgart high voltage atomic resolution TEM (JEM-ARM 1250, JEOL) operating at 1.25 MeV. This 0.12 nm point resolution microscope is equipped with a drift compensator, which enables highly stable working conditions at elevated temperatures up to 1000 ºC. All experimental work done for this study was conducted between 660-850 ºC (the melting point of Al is ~660 ºC) and recorded on negatives or on a real time TV-rate (25 frames per second) video camera. Experimental observations were obtained from pure single crystalline alumina (α-Al_2O_3, sapphire) specimens with different crystalline orientations. During irradiation with the high-energy electron beam the different threshold displacement energies of Al and oxygen mainly account for the selective displacement of Al ions. In-situ heating in high voltage TEM further accelerates the irradiation damage of alumina, resulting in the formation of liquid Al droplets.

Figure 1 shows the LS interfaces formed on different orientations of α-Al_2O_3; (a) (2-1-10), (b) (0001) and (c) (-2113) planes. While the (2-1-10) interface induces pronounced in-plane ordering of liquid Al atoms, no visible ordering is observed adjacent to the (-2113) interface. The liquid Al atoms in contact with α-Al_2O_3 (0001) interface are layered with a slight tendency of in-plane ordering for the first layer. These observations indicate that the atomic interaction of liquid Al atoms with sapphire depends critically on the atomic stacking of sapphire, which in turn results in the different atomic structures of LS interface. The interface-induced layering effect decays exponentially with the distance from the interface and eventually disappears.

In the case of LS (0001) interface, the oxygen adsorbed on the surface of liquid Al can be transported to the LS interface via the local oscillatory growth and dissolution reactions at an edge corner of the nanowire, particularly with the dynamic motion of the (0-114) facet (Figure 2). The interfacial diffusion of oxygen is constrained in-between the first ordered liquid Al layer and α-Al_2O_3 (0001) interface and forms an α-Al_2O_3 (0006) layer in its wake. Further details of oscillatory mass transport will be discussed.

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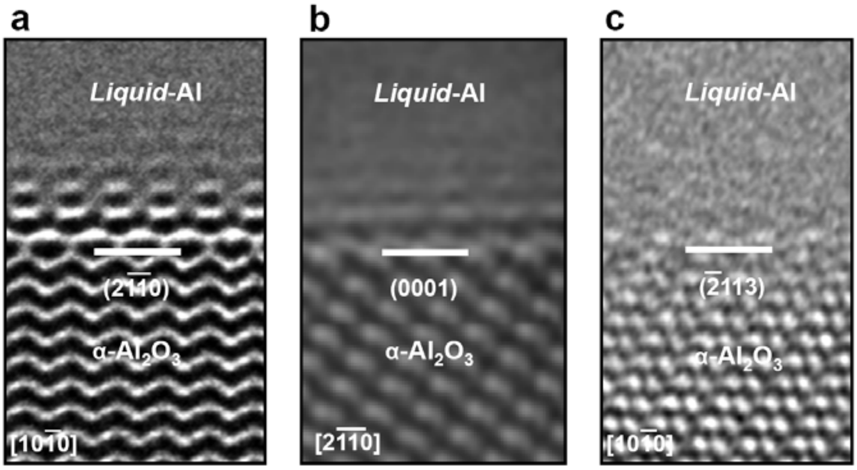

Figure 1. In-situ HRTEM images of liquid Al/sapphire LS interfaces with different orientations. (a) (2-1-10), (b) (0001) and (c) (-2113) planes.

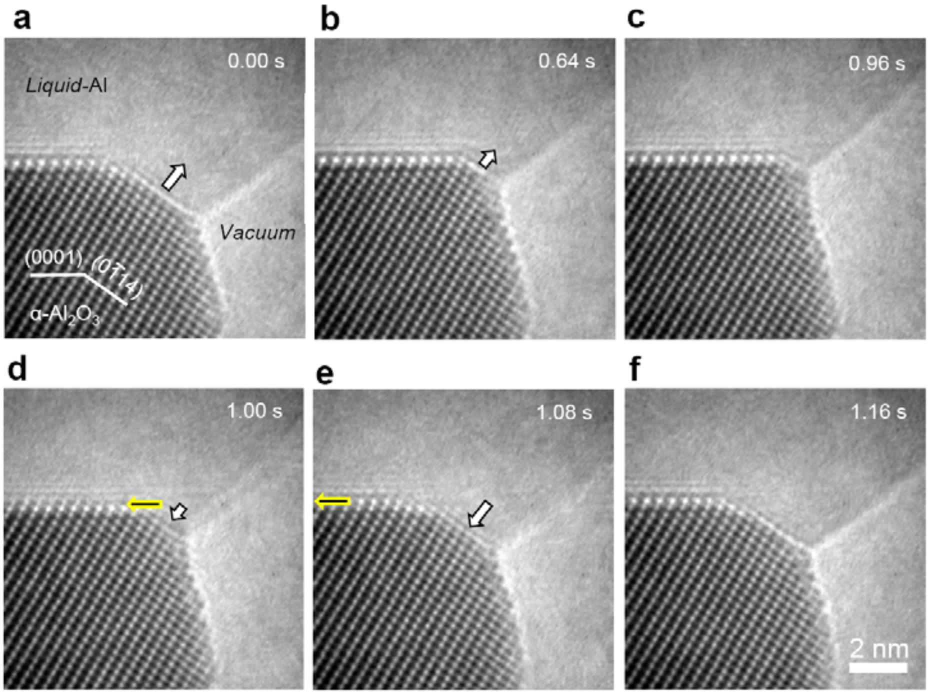

Figure 2. Diffusion of oxygen to the (0001) LS interface involving oscillatory growth and dissolution reactions at the triple junction. Video frame images in time intervals of (a) 0.00 s, (b) 0.64 s, (c) 0.96 s, (d) 1.00 s, (e) 1.04 s and (f) 1.16 s. The white arrow indicates the growth and dissolution motions of the (0-114) facet. The yellow arrow indicates the diffusion of oxygen along the liquid Al/sapphire (0001) interface.

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