Structure and Chemistry of Nanometer-Thick Intergranular Films at Au-Al_2O_3 Interfaces

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The existence of nanometer-thick amorphous equilibrium films at metal-ceramic interfaces has been experimentally verified for the Au-Al_2O_3 system [1]. The films were formed using a novel experimental approach, in which thin sputtered films of Au were dewetted on a sapphire substrate which was previously partially wetted with drops of anorthite glass (CaO-2SiO_2-Al_2O_3). The process resulted in sub-micron equilibrated Au particles residing on glass drops and on the sapphire substrate adjacent to the glass drops. The metal particles which formed on the glass drops 'sank' through the glass and reached the interface with the substrates.

Samples for transmission electron microscopy (TEM) were prepared from selected Au particles using a modified site-specific focused ion beam (FIB) lift-out method [2]. The interfaces between the Au particles and the sapphire substrates were characterized using a monochromated and aberration corrected TEM (Titan 80-300 S/TEM). The thickness of the films was determined from quantitative high resolution TEM (figure 1), and the composition was determined from combined energy filtered TEM (figure 2) and energy dispersive spectroscopy (EDS).

In addition, positive and relatively large Hamaker coefficients were calculated for the Au-film-Al_2O_3 interface, which indicates the existence of a stabilizing attractive van der Waals force, similar to intergranular films at grain boundaries in ceramics. Adjacent to the glass drops, a ~1nm thick surficial film was also detected on the (0001) surface of sapphire substrates partially wetted by anorthite glass. The refractive index required to stabilize the surface films, via a positive Hamaker constant, is explored through changes in the film composition. The relative interface energy of Au-Al_2O_3 and Au-Film-Al_2O_3 interfaces is explored via measured dihedral angles and use of the convoluted equilibrium crystal shape [3].

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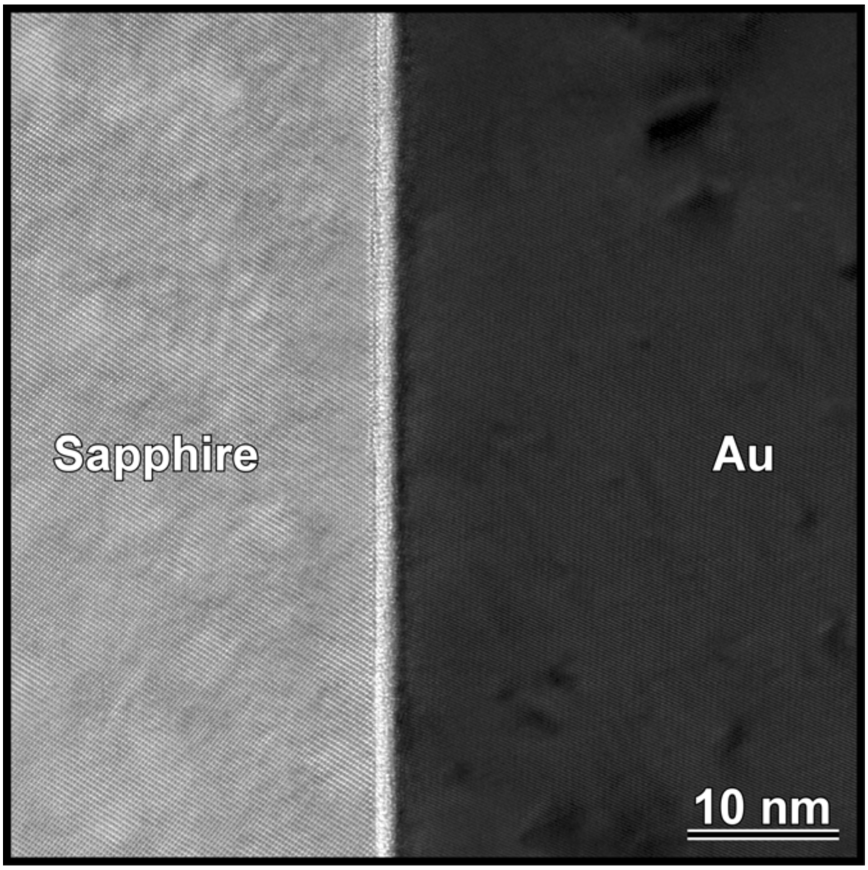

Figure 1. HRTEM (zero loss filtered) micrograph (Cs=-1.2µm) of the Au-sapphire interface. The sapphire was oriented in a $\left[1\bar{2}10\right]$ zone-axis before acquiring the micrograph, and the interface is parallel to the sapphire (0001) plane. There is no low-index orientation relationship due to the presence of the film at the interface, so only fringes are visible in the Au. The thickness of the interface was determined to be 1.09nm.

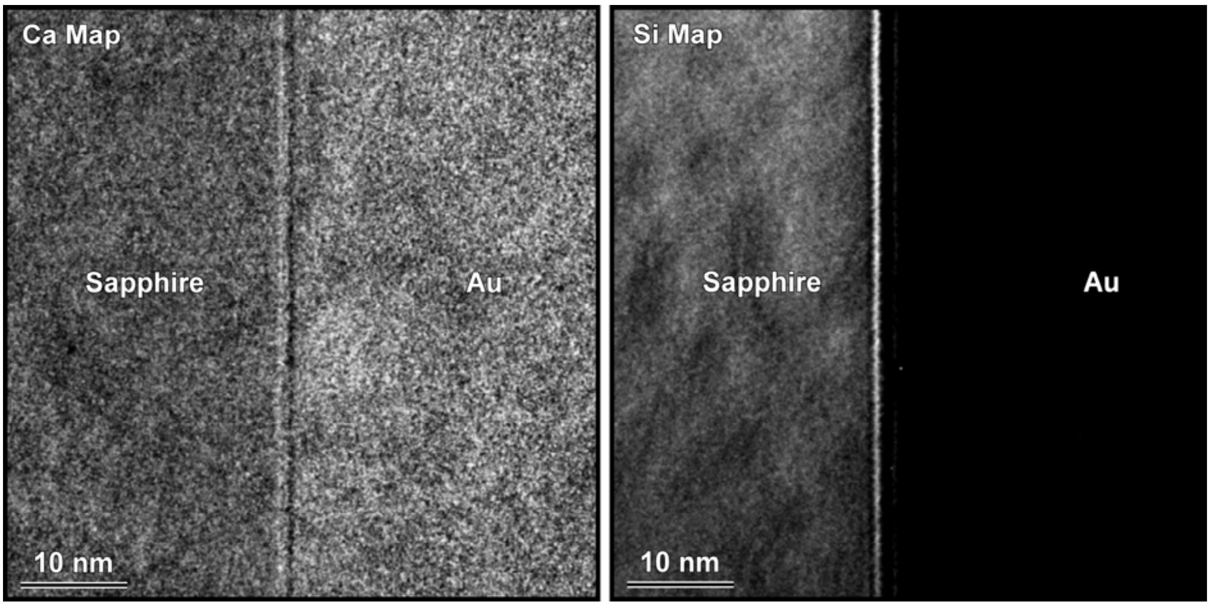

Figure 2. EFTEM images acquired using (a) the Ca L_3 (346 eV) and the (b) Si $L_{2,3}$ (99 eV) of the interface region, showing the existence of Ca and Si in the film.

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