Development of a Double-tilt Liquid Cell Holder for Zone-axis Incidence Imaging of FIB-prepared Single Crystal Samples

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Transmission electron microscopy (TEM) and scanning TEM (STEM) for imaging samples in a liquid cell use transmitted electrons passing through the top and bottom windows of the cell chips and liquid media. The window material of the cell chip fabricated via MEMS technology is popularly silicon nitride thin films with 30 to 50 nm in thickness, and a gap between two windows is usually several hundred nm, in which media and samples are enclosed [1]. Such thick windows and media worsen the resolution of TEM/STEM. Thus, it has been thought that atomic-resolution imaging of samples in a liquid cell is generally difficult. However, the contrast of atomic columns can be enhanced due to the electron channeling effect if the single crystal sample is aligned along a zone axis incidence, and this manner is common for conventional atomic-resolution STEM. In the present work, we developed a double-tilt liquid cell holder to image single crystal samples in a liquid on the zone-axis incident condition.

The sample was $SrTiO_3$ <001> lamella prepared with a focused ion beam (FIB) technique. This was prepared in an H-bar fabrication manner, and the center region was thinned to be less than 100 nm, and both sides were about 180 to 200 nm. There is an issue with transferring a FIB sample on a cell chip window using the in-situ transfer method with a dual-beam instrument equipped with a microprobe system because excessive Ga ion irradiation causes damage to a window during the transfer process. Ingenious procedures have been attempted to minimize Ga ion irradiation onto a cell chip window, followed by gas- and liquid-cell STEM observations [2,3]. Recently, the procedure of transferring FIB samples onto a window using an optical microscope and an electrically biased tungsten tip in the air has been proposed, which could fix the above issue [4]. We modified this method and applied it to the present work, e.g., a FIB-prepared $SrTiO_3$ <100> sample was picked-up and put onto a chip window at the desired position using a glass probe and an optical microscope as shown in Fig.1. The $SrTiO_3$ sample was immobile even when turning over the chip and when dropping a water droplet onto it, indicating good adhesivity between the sample and window. The chip was set in the double-tilt liquid cell vessel, and a pure water droplet of 0.1 µL was dropped onto it, followed by covering another chip. The enclosed chips form a fully sealed liquid volume housed in an on-chip fluidic chamber (see Fig.1(c)). Finally, it was fixed by a lid and screws to complete the assembly of the liquid cell holder, as shown in Fig.2.

Figure 3(a) shows an annular dark field (ADF) STEM image of the sample in the liquid cell, taken with an aberration-corrected STEM instrument, JEM-ARM200F. Oxygen signals from the area outside the sample in electron energy loss spectroscopy confirmed that a water layer existed there. Figure 3(b) shows an atomic-resolution ADF-STEM image from the thinnest area, acquired after aligning the sample orientation along the <100> zone axis. Sr and Ti-O atomic columns could be seen. Thus, it was demonstrated that electron channeling along several tens nm atomic columns resulted in high-contrast atomic images overcoming background from thick windows and media.

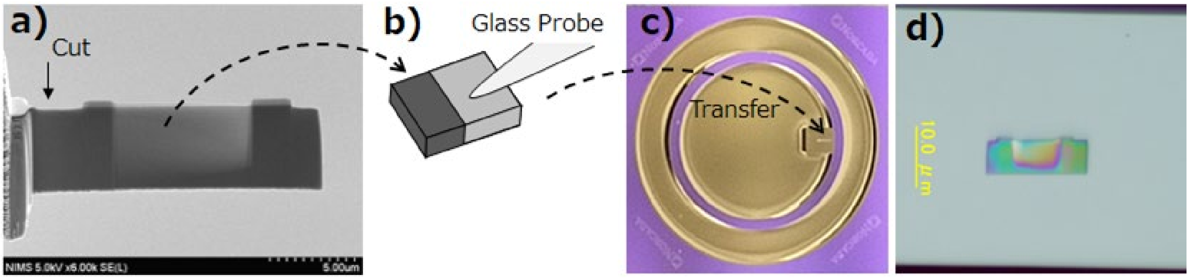

Fig. 1. (a) FIB-prepared $SrTiO_3$ <100> sample, (b) Sample pick-up using a glass probe, (c) Top-view of a liquid cell chip with an on-chip fluidic chamber, and (d) Sample on a chip window.

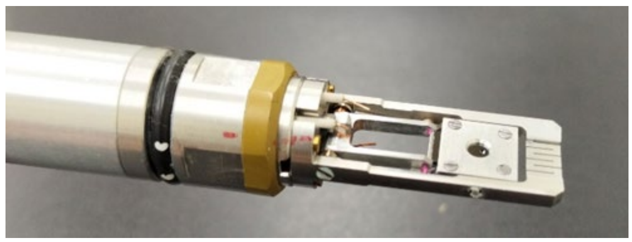

Fig. 2. Photo of the developed double-tilt liquid cell holder.

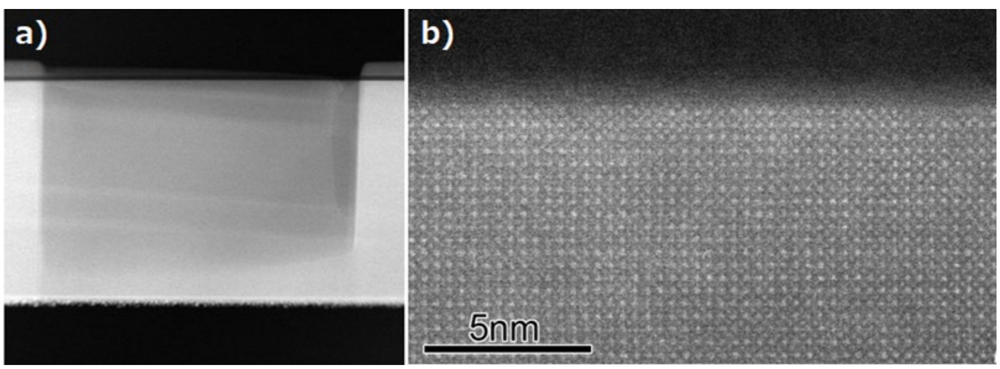

Fig. 3. (a) Low-magnification ADF-STEM image of $SrTiO_3$ <100> sample and (b) Magnified ADF-STEM image taken from a thin area of (a) on the zone-axis incidence condition (the off-line drift correction processed).

References:

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