Visualizing Bias-Dependent Changes in Ionic Liquid-$SrTiO_3$ Interface Structures by 3D scanning force microscopy

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Recently, electric double layer transistors (EDLTs) using ionic liquids (ILs) as gate insulators have attracted great attention. EDLTs allow to accumulate charges with a much higher density than conventional field-effect transistors by forming an electric double layer (EDL) at the IL/solid interface. To investigate its molecular-scale mechanism and device functionality, we previously constructed a simplified gold electrode model and performed 3D scanning force microscopy (3D-SFM) [1] imaging of an Au(111) surface in IL. These measurements successfully visualized bias-dependent changes in interfacial structures with subnanometer resolution [2]. However, in real EDLTs, transition metal oxides such as $SrTiO_3$ are used as the channel material and directly interface with the IL. The correlation between the structure of this IL/channel interface and device function has yet remained elusive. In particular, the large size and fabrication complexity of real EDLTs make them difficult to access using 3D-SFM.

To elucidate this correlation and overcome the measurement limitations, we developed a custom-made EDLT optimized for AFM observations. Specifically, we reduced the size of the $SrTiO_3$ substrate to 15 mm × 5 mm to fit within the AFM measurement space. The fabrication process included substrate annealing, resist coating, UV exposure using a mask aligner, Au deposition for electrode formation, followed by resist removal and development. This process yielded a compact EDLT device with exposed source, drain, gate electrodes, and a visible $SrTiO_3$ channel interface (Fig. 1a). Using this device, we performed 3D-SFM imaging of interfaces between DEME-TFSI and $SrTiO_3$ with variable bias voltage on gate electrode (Fig. (b)). The obtained images revealed multiple layer-like contrasts, with a spacing of approximately 0.8 nm, corresponding to the ion-pair size. Here, these layered structures are more distinct with a positive bias (Fig. 1c(i)) than with a negative bias (Fig. 1c(ii)), indicating a clear bias-dependent trend at the DEME-TFSI/$SrTiO_3$ interface. This trend is consistent with the expected behavior of EDLTs, in which a positive gate bias induces electron accumulation in the channel, whereas a negative gate bias leads to carrier depletion. We plan to perform detailed molecular dynamics (MD) simulations.

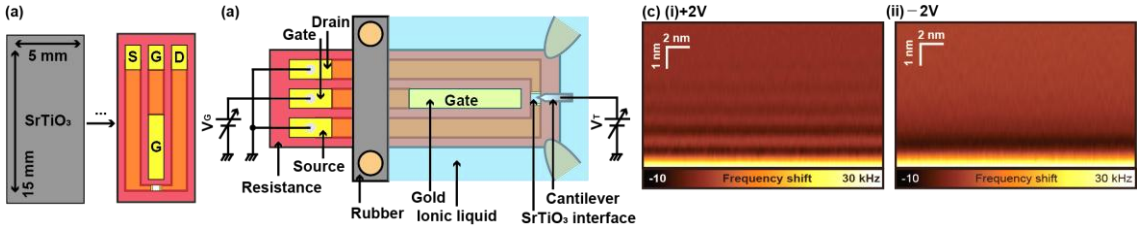

Fig. 1 (a)Before and after the EDLT fabrication. (b) Schematic of the measurement setup. (c) 3D-SFM images of DEME-TFSI-$SrTiO_3$ interface with (i) positive bias and (ii) negative bias.

References:

[1] T. Fukuma et al., Phys. Rev. Lett. 104 (2010) 016101.

[2] T. Ikarashi, et al., NC-AFM 2022.