Mapping nanoarchitecture of liquid-gel-solid interfaces: 3D nano-rheology microscopy nanoprobing from molecular layers to volume reconstruction

Oleg V. Kolosov[1,2], *Wenkai Wu*[3], *Yue Chen*[1,2,4]

[1]Department of Physics, Lancaster University, Lancaster, LA1 4YB, UK
[2]The Faraday Institution, Quad One, OX11 0RA, Didcot, UK
[3]College of Engineering, Swansea University, Bay Campus, Swansea SA18EN, UK
[4]Fujian Normal University, College of Physics and Energy, 350117, Fuzhou, China
E-mail of presenting author: o.kolosov@lancaster.ac.uk

One of the largest challenges for scanning probe microscopy (SPM) is exploiting its superior nanoscale to atomic resolution and SPM ability work in vacuum, air and liquid environments beyond the immediate surface of the sample.

A particular challenge is to study the soft matter surfaces (such as living cells or bacteria, polymer brushes or electrolyte-electrode interfaces in batteries) where the interfaces are not well defined and structures are truly three-dimensional. Here, we report novel 3D nano-rheology microscopy (3D-NRM) that uses a tiny (sub-nm to few nm) lateral dithering of the sharp SPM tip at kHz frequencies to probe the minute sample reaction forces. By mapping the increments of the real and imaginary components of these forces, while penetrating the soft interfacial layers, we obtain the true 3D nanoscale structure of sub–μm thick layers [1].

By combining 3D-NRM with the surface force-distance spectroscopy [2] (Fig 1d) in the *operando* electrochemical environment, we observed for the first time in real space the nanoscale dynamics of formation of the key solid electrolyte interphase (SEI) layer in Li-ion batteries starting from a few 0.1 nm thick electrical double layer to the full 3D nanostructured SEI (Fig 1e). We therefore were able to elucidate the key role of solvents in such formation and predict the conditions for building SEI for robust, safe and efficient Li-ion batteries.

The new 3D-NRM can provide unique opportunity for studies inorganic catalytic and separation processes, to explore biological interfaces probing bacterial and cell surfaces, and functional coating in the real space and appropriate operational environment.

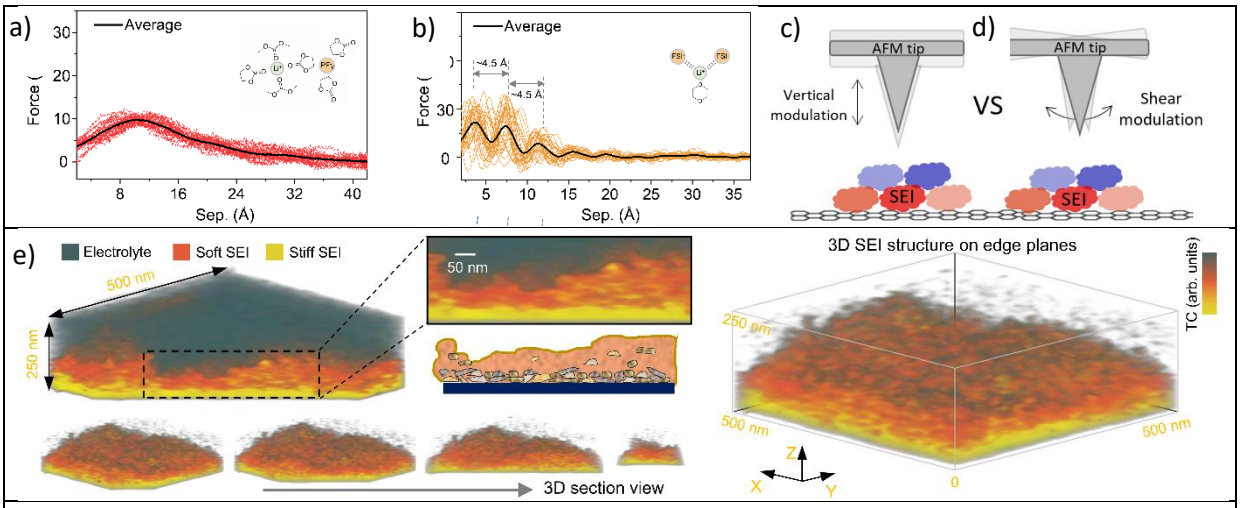

Figure1. a, b) Results of force-distance spectrocopy c) of the different electrolytes on the HOPG surface. e) 3D-NRM of the formed SEI in the Li-ion battery.

[1] Y Chen, W Wu, S Gonzalez-Munoz, L Forcieri, C Wells, SP Jarvis, F Wu, R Young, A Dey, M Isaacs, M Nagarathinam, RG Palgrave, N Tapia-Ruiz, OV Kolosov, *Nature Comm* 2023, 14, 1321.
[2] SJ O'Shea, ME Welland, JB Pethica, *Chem. Phys. Lett.* 1994, 223 (4), 336;