In-Situ AFM Investigation of Solid Electrolyte Interphase Formation and Failure Mechanisms in Lithium–Ion Batteries. THOMAS MUELLER, Bruker Nano Surfaces, RAVI KUMAR, ANTON TOKRANOV, Brown University, School of Engineering, TEDDY HUANG, CHUNZENG LI, Bruker Nano Surfaces, XINGCHENG XIAO, General Motors Global RD Center, BRIAN SHELDON, Brown University, School of Engineering — The formation and evolution of the solid electrolyte interphase (SEI) is critical for lifetime and performance of lithium-ion batteries (LIBs), particularly for LIBs with high energy density materials such as silicon. Si has almost ten times theoretical specific capacity vs graphite, but its volume changes during cycling (up to 400%) put enormous strains on the SEI layer, resulting in continuous capacity loss. In this study we report in situ atomic force microscopy (AFM) investigation on the formation and failure mechanisms of SEI layer using patterned Si island structures. Due to the shear lag effect, patterned Si islands go through lateral expansion and contraction, putting the SEI layer in tension and compression during lithiation and delithiation, respectively. Experimentally, we performed the studies in a glovebox with <1 ppm O2 and H2O, using PeakForce Tapping to image the extremely fragile SEI layer. We show for the first time the in operando cracking of SEI layer. To understand the mechanics of the SEI layer, the critical strain for cracking was derived from a progression of the AFM images. Our studies provide new insight into SEI formation, evolution and its mechanical response, and offer guidance to tailor passivation layers for optimal performance.