Abstract Submitted
for the MAR12 Meeting of
The American Physical Society

Scanning tunneling microscopy uncovers the mechanism of silicon oxidation in aqueous solutions MELISSA HINES, MARC FAGGIN, KUN BAO, ANKUSH GUPTA, BRANDON ALDINGER, Cornell University — Because of their immense technological importance, silicon oxidation reactions have been studied intensely for decades under a variety of conditions. However, the disordered nature of the reaction product, silicon oxide, makes these reactions notoriously difficult to understand. In this work, silicon oxidation is coupled with a subsequent etching reaction, allowing the oxidation reactions to literally write an atomic-scale record of their reactivity into the etched surface – a record that can be decoded into site-specific reaction rates, and thus chemical understanding, with the aid of simulations and infrared spectroscopy. This record overturns the long-standing and much-applied mechanism for the (low-temperature) oxidation of the technologically important face of silicon, Si(100), and shows that the unusually high reactivity of a previously unrecognized surface species leads to a self-propagating etching reaction that produces near-atomically-flat surfaces terminated by a single monolayer of hydrogen atoms. This finding shows that, contrary to expectation, the low-temperature oxidation of Si(100) is a highly site-specific reaction and suggests strategies for the uniform functionalization of the technologically relevant face of silicon by low-temperature reactions.

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Date submitted: 09 Nov 2011 Electronic form version 1.4