Abstract Submitted
for the MAR09 Meeting of
The American Physical Society

Link between optical second-harmonic and reflectance-anisotropy spectroscopy of stepped Si(001) surfaces ROBERT EHLERT, JINHEE KWON, MICHAEL C. DOWNER, University of Texas at Austin, Department of Physics, Austin, TX 78712-1081, USA. — Optical second-harmonic generation (SHG) and reflectance-anisotropy spectroscopy (RAS) are the two dominant non-invasive optical probes of electronic structure and chemical dynamics at surfaces, but underlying connections between these spectroscopies remain poorly understood. Here we combine spectroscopic SHG and RAS to characterize stepped Si(001) surfaces offcut toward [110] before and after dissociative adsorption of H_2 at the D_B step edges. Such stepped surfaces provide attractive templates for self-directed growth of nanoscale structures, while SHG/RAS provide non-invasive in-situ sensors to guide and interpret step edge chemistry. Our major finding is that the broad negative step-induced feature of the RA spectrum around 3eV, and the spectrum of the step induced third order Fourier component of the SHG azimuthal anisotropy, show strikingly similar spectral shapes and dependencies on H_2 adsorption, suggesting that these features share a common microscopic origin in the step edges. Separate analysis with a simplified bond hyperpolarizability model indicates that chemically active step dangling bonds are dominant contributors to both SHG and RAS, and that hydrogen termination of the step edges alters both by redistributing oscillator strength from the dangling bond to step back bonds.

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Date submitted: 10 Dec 2008 Electronic form version 1.4