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Spectroscopic second-harmonic generation and reflectance-anisotropy spectroscopy on reconstructed stepped Si(001) surfaces

ROBERT EHLERT, LOUCAS LOUMAKOS, MICHAEL C. DOWNER, University of Texas at Austin, Department of Physics, Austin, TX 78712-1081, USA — Spectroscopic 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]. Both the linear and nonlinear optical response of the clean surface are dominated by the large number of dangling bonds at the D_B steps and reconstructed terraces. Selective adsorption of H_2 at step edges and/or terraces allows us to isolate individual contributions and by using a simplified bond hyperpolarizability model (SBHM) identify their common microscopic origin. This ability to monitor surface chemistry on stepped silicon surfaces enables us to develop these methods as non-invasive, in-situ sensors to guide and interpret self-directed growth of nanoscale structures.

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