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Ultrafast Electron Dynamics of Silicon Surface States: Second-Harmonic Hole Burning Spectroscopy on Si(111)7x7[1] JOHN A. MCGUIRE, University of California, Berkeley, MARKUS B. RASCHKE, Max-Born-Institut fur Nichtlineare Optik und Kurzzeitspectroscopie D-12489 Berlin, Germany, Y.R. SHEN, University of California, Berkeley — We used second harmonic generation (SHG) as an all-optical, surface-specific probe of spectral hole-burning to measure the ultrafast dynamics of electronic excitations into the adatomU1 band of the Si(111)7x7 surface. The time-delayed SHG obtained with tunable ∼100 fs probe pulses was measured in response to the excitation of a ∼100 fs pump pulse at 1.54 eV. The transient holes induced by the pump corresponded to a homogeneous dephasing time as short as 15 fs. Carrier-carrier scattering was responsible for the linear dependence of the dephasing rate on pump fluence. The hole-burning spectra also reveal a strong coupling between the localized dangling-bond states and the surface phonon mode at 570 cm^{-1} associated with the adatoms. The technique can be extended to measurement of electron dynamics at any interface between two centrosymmetric media accessible to light.

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