We report on the first measurements of the vibrational lifetimes of hydrogen- and deuterium-related stretch modes in crystalline silicon by high-resolution infrared absorption spectroscopy and pump-probe transient bleaching technique. The lifetimes are found to be extremely dependent on the defect structure, ranging from 2 to 340 ps. Vibrational lifetimes have been obtained in the temperature range 4 - 300 K and will be discussed based on phonon-phonon interactions. Hydrogen bonds strongly to virtually any imperfections such as intrinsic defects, impurities, surfaces, and interfaces in crystalline silicon. These hydrogen-containing defects give rise to absorption lines in the mid-infrared region, which originate from excitations of local vibrational stretch modes. The most prominent lines have been identified as hydrogen-decorated vacancies ($V_mH_n$), self-interstitials (IH$_2$), and the H$_2^+$ defect. Hydrogen implanted into silicon at 80 K gives rise to an extremely intense absorption line at 1998 cm$^{-1}$ previously identified as the positive charge state of hydrogen located at the bond-center site in an otherwise perfect silicon crystal. Samples containing hydrogen (deuterium) are produced by multiple-energy implantation of protons (deuterons) into silicon samples. The vibrational lifetimes of the hydrogen and deuterium related modes are determined by measuring the recovery of the transient bleaching induced by intense picosecond infrared pulses from the Jefferson Lab. Free-Electron Laser (FEL) [1]. These lifetimes are found to be much shorter than Si-H stretch modes on hydrogen-terminated Si surfaces (>1000 ps) indicating that these modes strongly couple to optical phonons. It is shown that the width of the absorption lines are determined by the vibrational lifetime for $T < 40$ K, whereas dephasing determines the line shape for $T > 40$ K. Against conventional wisdom, we find that lifetimes of Si-D modes typically are longer than for the pertinent Si-H modes. The potential implications of the results on the physics of electronic device degradation are discussed. This work is supported in part by DOE (DE-FG02-99ER45781), NSF (DMR-00-76027) and the Jeffress Foundation (J-545).

References