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High-Resolution Spectroscopy with a Free-Electron Laser: Vibrational Lifetimes of Hydrogen-related Defects in Silicon[1]

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Gunter Luepke, Department of Applied Science, The College of William and Mary, Williamsburg, VA 23187 Vibrational lifetimes of hydrogen- and deuterium-related bending and stretching modes in crystalline silicon are measured by high-resolution infrared absorption spectroscopy and pump-probe transient bleaching technique using the Jefferson Lab. Free-Electron Laser. We find that the vibrational lifetimes of the bending modes follow a universal frequency-gap law, i.e., the decay time increases exponentially with increasing decay order, with values ranging from 1 ps for a one-phonon process to 265 ps for a four-phonon process. The temperature dependence of the lifetime shows that the bending mode decays by lowest-order multi-phonon process. In contrast, the lifetimes of the stretching modes are found to be extremely dependent on the defect structure, ranging from 2 to 295 ps. Against conventional wisdom, we find that lifetimes of Si-D stretch modes typically are longer than for the corresponding Si-H modes. Our results provide new insights into vibrational decay and the giant isotope effect of hydrogen in semiconductor systems. The potential implications of the results on the physics of electronic device degradation are discussed.

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