

Abstract Submitted
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Theory of the lifetime of adsorbate vibrations on semiconductor surfaces PETER KRATZER, SUNG SAKONG, University Duisburg-Essen — On semiconductor surfaces, the vibrational lifetime of covalently bonded adsorbates is rather long (nanoseconds or more) since the band gap precludes electronic dissipation. Due to the quantum nature of vibrational states, such slow relaxation is expected, as the large quantum of the bond stretching must be converted into several smaller (phononic or vibronic) quanta by a high-order process. We use density functional theory calculations to map out the high-dimensional potential energy surface governing the anharmonic coupling of the stretching to the local bending and shift modes, while the coupling to the substrate phonons is treated perturbatively. Applying our method to the vibrational lifetime of CO on Si(100), we find that the CO stretching relaxes predominantly via an intermediate state consisting of four shift and/or bending quanta and one phonon. Good agreement with the measured lifetime of 2.3 ns at $T = 100$ K is achieved. As a second application, we elucidate the role of intermediate vibrational states in the relaxation of the stretching vibration at Ge(100):H surfaces. For Ge surface dimers saturated by one H and one D atom, the lifetime of the Ge–H stretching vibration is up to five times shorter and less temperature-dependent than in Ge dimers homogeneously saturated by H. Our analysis shows that the symmetry breaking associated with the isotope mixture opens up additional relaxation channels.

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