Ultrafast Time Resolved Electron Diffraction of Adsorbate Dynamics on Silicon Surfaces

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Ultrafast time resolved electron diffraction is an excellent technique to study dynamic processes of surfaces like phase transitions and vibrational mode coupling on a picosecond timescale after excitation by a fs laser pulse. The laser energy will excite the electron system and heat the topmost atomic layers by electron-phonon coupling. In our experiment surface sensitivity is obtained by a RHEED (reflection high energy electron diffraction)-geometry [1].

The transient dynamics of the metallic $(\sqrt{3}\times\sqrt{3})$ and the $(\sqrt{3}\times\sqrt{7})$-Pb reconstructions on Si(111) has been studied in a laser-pump and electron-probe setup. The de-excitation and energy dissipation into the substrate has been measured through the transient surface temperature using the Debye Waller effect on the diffraction pattern. We observe two different time constants of 100 ps and 2800 ps for the de-excitation of the vibrational modes for both systems. These long time constants can be explained by the huge difference in mass of Si and Pb atoms which prevents effective coupling of the Pb vibrational modes to the phonon bath in the Si substrate.

In order to study the dynamics of strongly driven phase transitions at surfaces far away from thermal equilibrium we performed time resolved diffraction on the Peierls like phase transition from a (8 × "2") to a (4 × 1) reconstruction of a Indium terminated Si(111) surface upon laser excitation [2]. This phase transition is observed by preparing the (8 × "2") well below 90 K and excitation of the surface with a fs-laserpulse. After excitation the (8×"2")-diffraction spots instantaneously disappears, while the intensity of the (4 × 1) spots increases. This increase of the (4 × 1) spot intensity excludes an explanation by the Debye-Waller-Effect and is evidence for a true structural phase transition at a surface.

[1] A. Janzen et al., Rev. Sci. Inst. **78**, 013906 (2007).

[2] S. Möllenbeck et al., MRS-Proceedings (accepted) (2010).