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Probing phase transitions at surfaces with ultrafast electron diffraction

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The multitude of possible processes that can occur at surfaces cover many orders of magnitude in the time domain. While large scale growth and structure formation happens on a timescale of minutes and seconds, diffusion is already much faster, but can still be observed by electron microscopy. Many other processes as chemical reactions, phonon dynamics, or phase transitions, however, take place on the femto- and picosecond timescale and are yet way to fast for imaging techniques. In order to study such ultrafast processes at surfaces we have combined modern surface science techniques with fs laser pulses in a pump probe scheme. We use a RHEED setup with grazing incident electrons of 7 - 30 keV to ensure surface sensitivity. In order to overcome the velocity mismatch between light and electrons a tilted pulse front scheme is used to achieve a time resolution of less than 2 ps. The sample is excited with 800 nm photons with a pulse energy of 0.5 mJ at 5 kHz repetition rate. The huge potential of this technique for the study of transient surface phenomena is demonstrated with the non-equilibrium dynamics of the In induced c(8x2) reconstruction on Si(111). This surface exhibits a Peierls-like phase transition at 100 K from a c(8x2) groundstate, which is accompanied by the formation of a charge density wave (CDW), to (4x1) excited state. Upon excitation by the fs-laser pulse this structural phase transition is driven into the excited (4x1) state at a sample temperature of 20 K. The surface is only excited electronically, the CDW is lifted by photo doping and the surface remains up to 500 ps in a super cooled excited (4x1) state. Relaxation into the c(8x2) groundstate happens delayed through the nucleation of the c(8x2) at defects which triggers a 1-dim. recrystallisation front which propagates with the velocity of sound. Utilizing the Debye Waller effect, the excitation, conversion and relaxation of vibrational excitations in monolayer adsorbate systems like the Pb induced HIC ($\sqrt{3}$x$\sqrt{3}$) phase on Si(111) was studied. Initially only a high frequency optical mode with an amplitude parallel to the surface is excited. Subsequently, this mode decays into low frequency acoustic modes with an amplitude vertical to the surface which, however, do not couple to the phonons of the Si substrate and survive for many nanoseconds.