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Second Order Phase Transition in Si(001) & Advances in the tr-RHEED Experimental Setup

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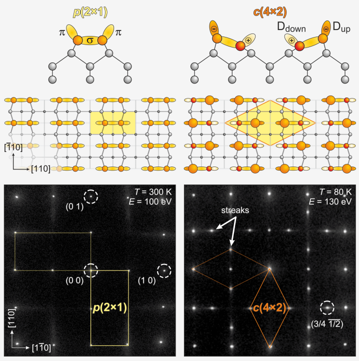
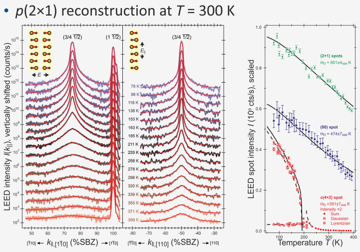
Second Order Phase Transition of Si Dimers on the Si(001) Surface



Continous Order-Disorder Transition of Buckled Dimers on Si(001)-$c(4\times2)$

- Buckled dimers on Si(001) form $c(4\times2)$ reconstruction at low temperatures
- Thermally activated dimer flipping causes disorder for $T > 200$ K through
  - Generation of 0D domain boundaries
  - Phase shifts of dimer rows
  - Broadening of $c(4\times2)$ diffraction spots
- Anisotropic coupling between dimers along and across rows
- $p(2\times1)$ reconstruction at $T = 300$ K



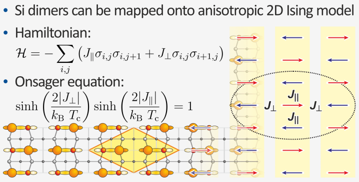
Thermally Induced Second Order Phase Transition

- Si dimers can be mapped onto anisotropic 2D Ising model
- Hamiltonian:

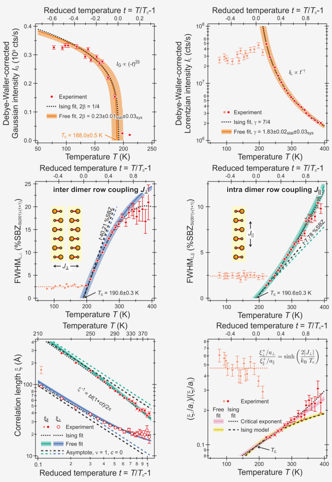
$$\mathcal{H} = -\sum_{i,j}\left(J_{\parallel}\sigma_{i,j}\sigma_{i,j+1} + J_{\perp}\sigma_{i,j}\sigma_{i+1,j}\right)$$

- Onsager equation:

$$\sinh\left(\frac{2|J_{\perp}|}{k_B\ T_c}\right)\sinh\left(\frac{2|J_{\parallel}|}{k_B\ T_c}\right) = 1$$

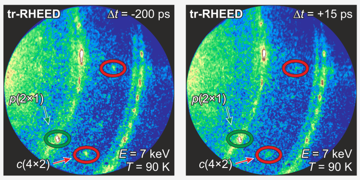


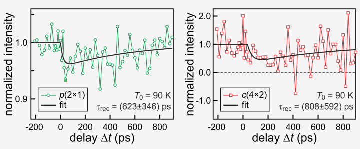
- Si(001)-$c(4\times2)/p(2\times1)$ phase transition at $T_c = 190.6$ K
- Determination of critical exponents by spot profile analysis
- Correlation length ratio above T_c determines dimer coupling
- Coupling energies of dimers:
  - Along dimer rows $J_{\parallel} = -24.9 \pm 1.3$ meV
  - Across to dimer rows $J_{\perp} = -0.8 \pm 0.1$ meV



Non-Equilibrium Dynamics of Order-Disorder Transition Triggered by fs-Laser Pulses

- Impulsive lifting of Jahn-Teller distortion
  - Ultrafast dynamics of initial motion
  - Thermal or non-thermal excitation?
  - High excitation fluences up to 14 mJ/cm^2
  - fs-pulse absorption only at surface states, Si bulk (almost) transparent
- Recovery of ground state
  - Nucleation and coarsening of ordered domains
  - Decoupling of ordering along and across dimer rows
  - Kibble-Żurek mechanism
  - Kinetically limited critical slow down
- Finite size effects by vicinality of surface?
- Dependence of transition on
  - Base temperature T_0?
  - Laser fluence Φ?
  - Photon energy $h\nu$?



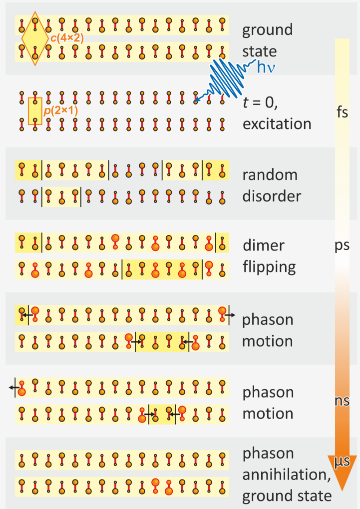


S. Möllenbeck *et al.*, MRS Proceedings **1230**, MM03-09 (2009)

Phasons: Creation, Motion & Annihilation

- Topologically protected 0D domain boundaries along dimer rows
- Nucleation and growth of ordered domains dependent on diffusion, annihilation and collective motion of phasons
  - Nucleation of 1D domains of alternately buckled dimers by dimer flipping
  - Coarsening of domains through diffusion and pair-wise annihilation of 0D domain boundaries (phasons)

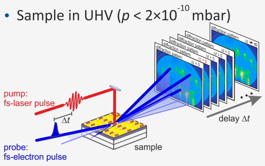
Excitation of Disorder Through fs-Laser Pulses and Recovery of Ground State



Advances of the Optical Setup and Implementation of the New Single Electron Detector

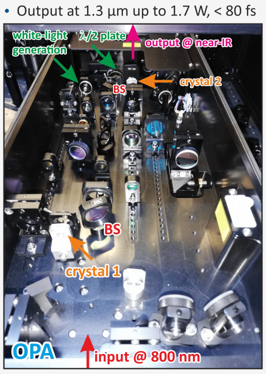
Ultrafast Electron Diffraction at Surfaces

- Femtosecond reflection high energy electron diffraction (fs-RHEED)
- Surface sensitivity through grazing incidence
- Temporal resolution of 330 fs meets the relevant time scales
- Laser pump - electron probe setup
- Sample in UHV ($p < 2\times10^{-10}$ mbar)



Optical Parametric Amplification

- Two-stage parametric amplifier of white-light continuum (*Light Conversion TOPAS-Prime*)
- Variable photon wavelength in near IR: signal (1.16-1.6 µm), idler (1.6-2.6 µm)
- Output at 1.3 µm up to 1.7 W, < 80 fs

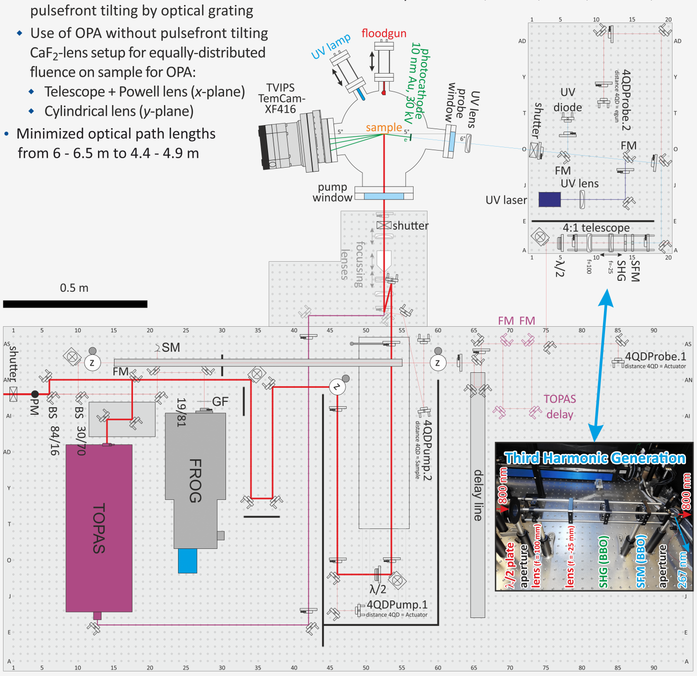


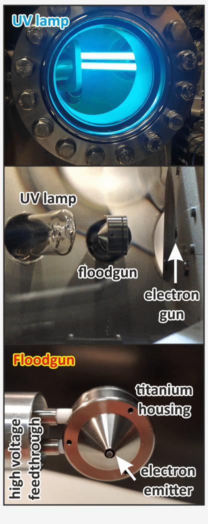
Simplified Optical Setup

- Output from regenerative amplifier (*Coherent Legend*): 800 nm, 80 fs, 1 mJ, 5 W, 5 kHz
- Power distribution: 84% for optical pump, 5% for probe, 2% for spectrometer & FROG, 9% for TROS
- Selection of optical pump: either at 800 nm or at longer wavelength in near-IR via OPA
  - Optimal temporal resolution at 800 nm using pulsefront tilting by optical grating
  - Use of OPA without pulsefront tilting CaF_2-lens setup for equally-distributed fluence on sample for OPA:
    - Telescope + Powell lens (*x*-plane)
    - Cylindrical lens (*y*-plane)
- Minimized optical path lengths from 6 - 6.5 m to 4.4 - 4.9 m

Compact Third Harmonic Generation & UV Laser

- Electron generation from UV light at $h\nu = 4.65$ eV in Au film photocathode
- Compactified THG setup to fit into breadboard on RHEED table
- Easy alignment of optics in cage system
- UV laser (*BrightMicrolaser P4*) for long temporal delays: 236.5 nm, 0.94 ns, 180 nJ, 0.94 mW, 5 kHz





Next Generation Electron Detection:

- New direct detection of diffracted electrons:
  - *TVIPS TemCam XF416* CMOS detector
  - Single electron detection
  - 63.5 × 63.5 mm^2, 4096 × 4096 pixels
  - Cooled to 0°C by Peltier elements
  - Non-bakeable detector mounted on custom-made gate valve (*VAT*)
  - Chiller (*Van Der Heijden Kühlmobil*)
- UV lamp (*RBD Instruments UVB-100*):
  - Desorption of adsorbates from detector surface after mounting
  - Improvement of UHV conditions
- Custom-made electron floodgun by *Th. Duden*:
  - Homogeneous illumination of complete detector area for flatfields
  - Electron emission up to 20 keV
  - Y_2O_3-coated Iridium cathode (*Kimball Physics ES-529*)
- Proof of principle at $E_e = 20$ keV: Epitaxial monolayer graphene on SiC(0001) at room temperature