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Electron-Phonon coupling in thin Pb films on Si(111) from the lattice's perspective

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SFB 1242

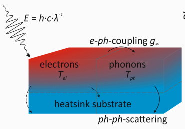
DFG Collaborative Research Centre 1242: **Non-Equilibrium Dynamics of Condensed Matter in the Time Domain**

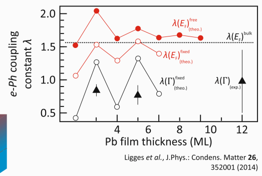
Motivation

- Understanding of the fundamental mechanisms of energy transfer between electron and phonon system after optical excitation in the time domain, from the lattices perspective

Special focus on two aspects:

- Initial dynamics after optical excitation
- Quantum well states of Pb/Si(111)

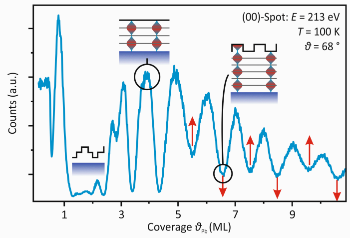


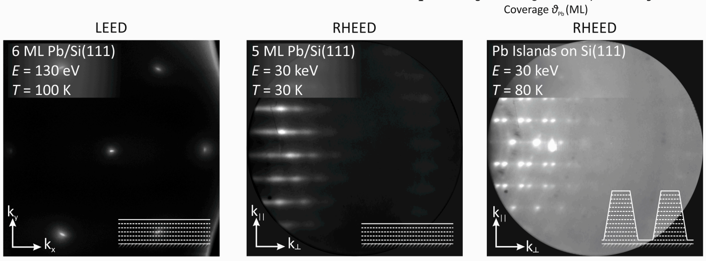


Ligges et al., J.Phys.: Condens. Matter **26**, 352001 (2014)

Material System and Quantum Size Effect

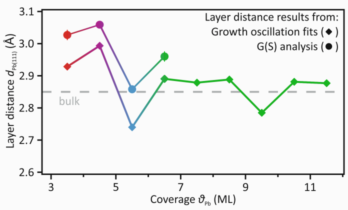
- Growth of epitaxial thin Pb-films on Si(111) possible at low sample temperatures ($T \leq 100$ K)
- Control of surface quality by using a Si-terminating template layer of Pb (e.g. β-($\sqrt{3} \times \sqrt{3}$))
- Growth observable *in-situ* by intensity oscillations of diffraction spots
- Alternating intensity of minima (s. red arrows)

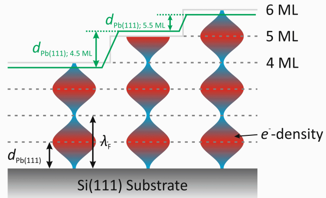




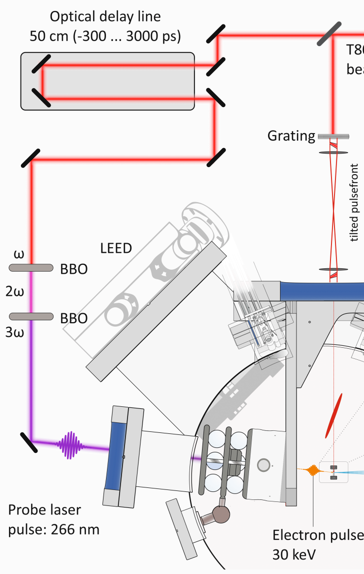
Quantum Size Effect

- Experiments show clear thickness dependence of layer spacing $d_{Pb(111)}$
- Finite thickness of films leads to standing waves in electron density





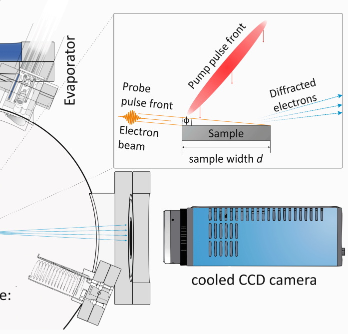
Experimental Setup



regenerative amplifier output: 800 nm, 80 fs, 5 kHz, 1 mJ

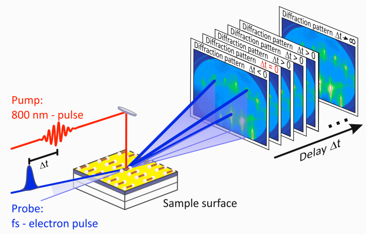
T80:R20 beamsplitter

- Probing: 30 keV electrons, 1-5° gracing incidence
- Temporal width: > 300 fs, $N \geq 500\ e^-$, E_{ext} = 7.5 kV/mm, ξ_{coh} = 54 nm
- Pumping: 80 fs IR at 0...8 mJ/cm²
- UHV chamber with $p \approx 2\cdot10^{-10}$ mbar, 30 K < T < 1550 K, *in-situ* sample preparation



Time-Resolved Electron Diffraction

- Time-resolved measurements using electron diffraction at the surface (tr-RHEED) in a pump-probe setup
- Temporal resolution achieved with current setup $\tau_{FWHM} \geq$ 350 fs
- Analysis of transient diffraction pattern
- Optical excitation of electron subsystem
- Excitation of phonon system via electron-phonon coupling

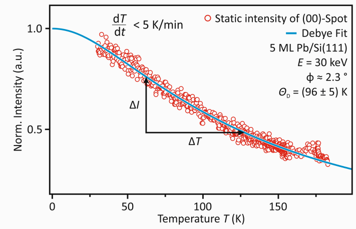


Debye-Waller effect

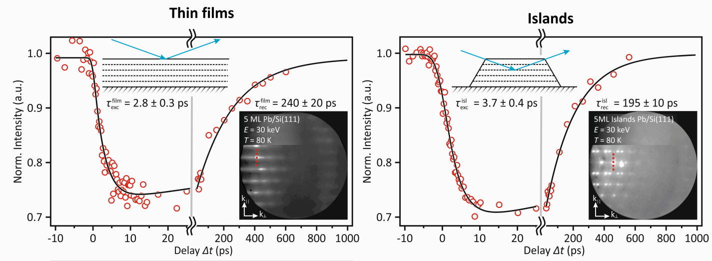
- Excitation of phonon system leads to incoherent motion of surface atoms → loss of diffraction intensity:

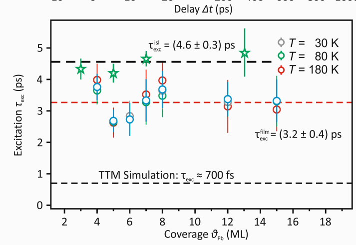
$$I_{DBW} \approx A \exp(-\alpha\ T_{Ph}(t))$$

- Increase of lattice temperature observable in transient diffraction intensity



Experimental results: transient intensity





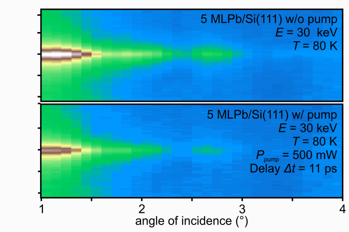
- Experimental lattice excitation is significantly slower than expected:
- Excitation of Pb/Si(111) surface almost independent of film thickness at τ^{film}_{exc} = 3.2 ± 0.4 ps
- Surprisingly: Lattice excitation in islands roughly 50% slower τ^{isl}_{exc} = 4.6 ± 0.3 ps

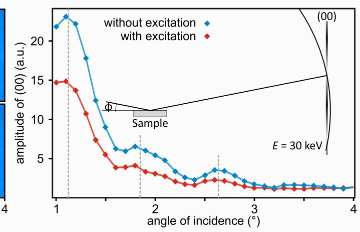
Comparison with Two Temperature Model

$$C_{el}\ \partial T_{el}/\partial t = -g_{\infty}(T_{el}-T_{ph}) \quad \& \quad C_{ph}\ \partial T_{Ph}/\partial t = g_{\infty}(T_{el}-T_{ph}) - \sigma_{TBC}(T_{Ph}-T_{Substrate})$$

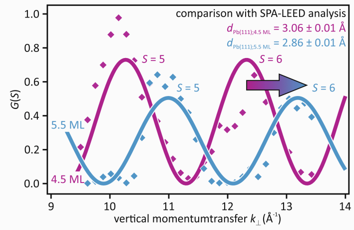
- Heat transport within film is negligible
- Numerical simulation with Pb's specific constants
- Expected time constant of excitation $\tau^{TTM}_{exc} \approx$ 0.7 ps
- g_{∞} is expected to show bilayer oscillation due to quantum size effects (see Motivation)

Preliminary results: rocking curve at ϑ_{Pb} = 5 ML



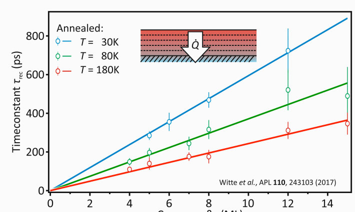


- 5 ML of Pb on Si(111) exhibits increased layer spacing
- Layer spacing relaxes to bulk condition at 6 ML film thickness
- Checking layer spacing in RHEED: does d_{111} change after optical excitation?



Heat transfer across surface boundary

- Cooling of the lattice via phonon transmission across the Pb/Si interface
- $\dot{Q} = \sigma_{TBC}\,\Delta T$ σ_{TBC}: Thermal Boundary Conductance [W/m²K]
- Data well described with $\sigma_{TBC} = c_V\ \rho\ (\partial/\partial d_{Pb}\ \tau_{rec}(d_{Pb}))^{-1}$
- Thermal Boundary Conductance depends on substrate temperature
- Used to doublecheck layer thickness or determination of island height



Witte et al., APL **110**, 243103 (2017)