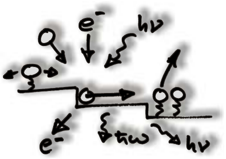
UNIVERSITÄT DUISBURG ESSEN

SFB 616

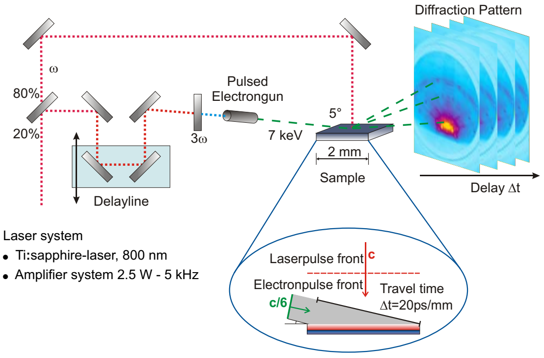
Ultra-Fast Time Resolved Electron Diffraction at Surfaces

S. Möllenbeck, A. Hanisch, T. Pelka, P. Schneider,
B. Krenzer, M. Horn-von Hoegen



B2

Introduction



Laser system
- Ti:sapphire-laser, 800 nm
- Amplifier system 2.5 W - 5 kHz

A possibility to study the structural dynamics of surfaces directly is time resolved electron diffraction. The major difficulty when doing time resolved surface sensitive diffraction experiments is to achieve the shortest possible time resolution and to stay surface sensitiv. The solution is to use a time resolved RHEED (Reflection High Energy Electron Diffraction) setup. Therefore we combined a UHV RHEED (diffraction at clean, well defined surfaces) with a pump probe setup (fs laser-system).
But due to the different incidence angles of electron and laser beam and the different propagation velocities the velocity mismatch is the limiting factor of the time resolution.

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

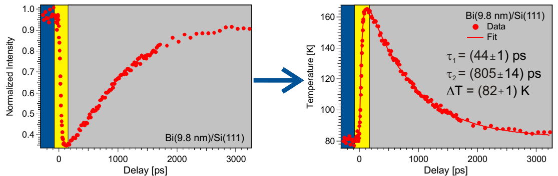
Measurement

After heating the sample the intensity of the diffraction pattern decreases. This intensity change can be converted in a surface temperature evolution by using the Debye-Waller Effect. The Surface Debye Temperature can be determined in a static experiment.
The transient temperature evolution shows three different regions. They all can be well described by one empiric fit function.

$$I(T) = I_0 e^{-2M} = I_0 e^{-\frac{1}{3}\left|\vec{k}\right|^2 \langle \vec{u}^2 \rangle}$$

$$\langle \vec{u}^2 \rangle = \frac{3\hbar^2}{m k_B \theta^2_{D,Surf}} T$$

	Bulk Θ_D [K]	Surface Θ_D [K]
Bi	119	48
Pb	90	49
In	111	49
Si	645	458

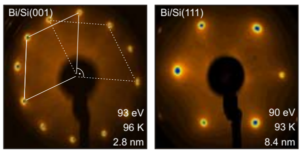


$$T(t) = \Theta(t)\Delta T \left(1 - \boxed{e^{-t/\tau_1}}\right) \boxed{e^{-t/\tau_2}} + T_0$$

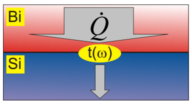
B. Krenzer et al., New J. Phys. **8**, (2006),
A. Janzen et al., Surf. Sci. **600** (2006).

Thin Films

Thin Bi films on Si(001) and Si(111) have been investigated.
On Si(001) two different recipes were used:
a) Bi deposition on Si(001)-c(4x2) at 300 K
b) Bi deposition (2.8 nm) on Si(001) at 150 K annealed at 420 K and additional deposition at 400 K.
With recipe b) it is possible to prepare smoth, closed Bi-film with thicknesses < 3 nm.
On Si(111) Bi was deposited (2.8 nm) on Si(111)-(7x7) at 150K, annealed at 420 K and additional deposited at 400 K.



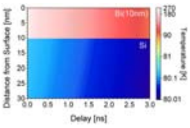
G. Jnawali et al., Phys. Rev. B **74**, 195340 (2006),
T. Nagao et al., Surf. Sci. **590**, L247 (2005).



The observed decayprocess is much slower in comparison to the heat diffusion in a Bi- single crystal. The interface between Bi and Si forms a barrier for thermal diffusion.

$$Cd\frac{\partial T(t)}{\partial t} = -\sigma_K (T_f(t) - T_s(t))$$

$$\sigma_K = \frac{1}{2}\int_0^\infty C(\omega,T)\,\langle v_z(\omega)\rangle\langle t(\omega)\rangle\,d\omega$$



E.T. Swartz, R.O. Pohl, Rev. Mod. Phys. **61**, (1989).

B. Krenzer et al., J. Nanomat. **2008**, 590609 (2008).

Acoustic Mismatch Model

The transmission probability can be calculated by a theoretical model. Therein phonons are described as elastic waves. In analogy to optics the „Fresnel"-equations or "Snells Law" can be used to describe total internal reflection of the phonons at the interface. Due to the large difference in the speed of sounds only a small amount of phonons can cross the interface.

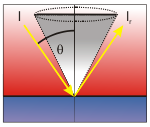
critical cone of total reflection

$\sin\theta_{max} = c_{Bi}/c_{Si} \Rightarrow \theta_{max} = 13.5°$

$c_{Si,t} = 5845$ m/s $c_{Si,l} = 8433$ m/s

$c_{Bi,t} = 1074$ m/s $c_{Bi,l} = 1972$ m/s

$\Rightarrow$ **$\sigma_K = 1400$ W/(cm²K)**



J. Miklowitz, Applied Mathematics and Mechanics, 2nd Ed., (1980).

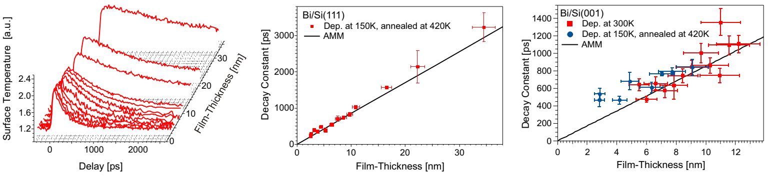
B. Krenzer et al., New J. Phys. **8**, (2006).

Thickness Dependence

Using the thickness dependence of the decay constant τ_2 we can compare the theoretical Thermal Boundary Conductance of the AMM (calculated using bulk values) with the experimental results.

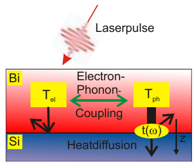
$$\tau_2 = \frac{C}{\sigma_K} \cdot d$$

A. Hanisch et al., Phys. Rev. B **77**, 125410.



Two Temperature Model

The heating of the sample can be described with the Two Temperature Model (2TM).
In the 2TM the electrons and the phonons are treated as two systems, linked via the Electron-Phonon-Coupling.



$$C_{el}(T_{el})\frac{\partial}{\partial t}T_{el} \overset{\text{Electrons}}{=} \kappa_{el}\frac{\partial^2}{\partial z^2}T_{el} - g_\infty(T_{el}-T_{ph}) + A(z,t)$$

$$C_{ph}(T_{ph})\frac{\partial}{\partial t}T_{ph} \overset{\text{Phonons}}{=} \kappa_{ph}\frac{\partial^2}{\partial z^2}T_{ph} + g_\infty(T_{el}-T_{ph})$$

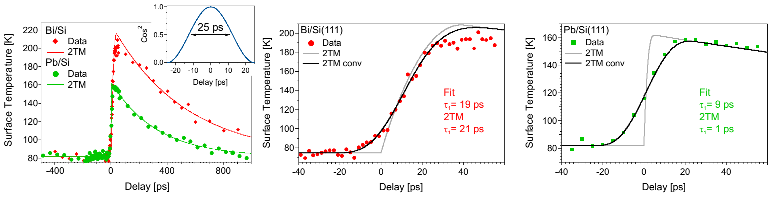
With the simple assumption

$(T_{el}-T_{ph}) \ll T_{ph},\ T_{ph} \gg \theta_D,$ $$g_\infty = \frac{3\hbar}{\pi k_B}\gamma\lambda\left\langle\omega^2\right\rangle$$

	Bismuth (Bi)	Lead (Pb)
γ	0.372 J/(m³K²)	163.1 J/(m³K²)
λ	0.2 – 0.4	1.45
$\langle\omega^2\rangle$	160 meV²	31 meV²
g_∞	**0.0303·10¹⁶ W/(m³K)**	**12.37 ·10¹⁶ W/(m³K)**

Ph. Hoffmann, Prog. Surf. Sci. **81**, (2006),
S. D. Brorson, Phys. Rev. Lett. **64**, (1990),
Ch. Kittel, Oldenbourg 12th Ed. (1999).

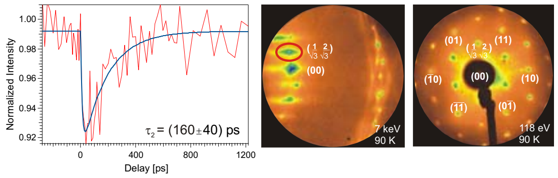
the 2TM for Bi and Pb can be calculated and compared to the experimental results.
To take the temporal resolution into account the 2TM data were convoluted with cos² with a width of 25 ps.
This model can reproduce the measured data for the thin Bi-films.
For the Pb-films the temporal resolution is insufficient.



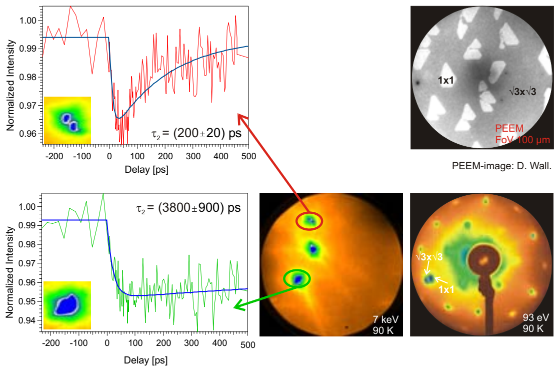
Monolayers

Lead (Debye Waller)

Thin Pb films were prepared on Si(111)-(7x7) at 100 K and slowly annealed to RT. By desorption at 500 K the coverage was reduced to a (√3x√3) reconstructed Pb Monolayer. The measured time constant τ_2 for the ML-system is 150 to 200 ps.



By deposition of Pb at RT the formation of a wettinglayer and large flat islands takes place, shown in the PEEM-image. The wettinglayer is again (√3x√3) reconstructed, the islands (1x1) with a larger lattice constant.
In a time resolved measurement of this heterosystem two different temporal behaviours can be observed simultaneously. For the wettinglayer the time constant τ_2 is again between 150 and 200 ps, whereas the islands show a time constant τ_2 of around 3500 ps.

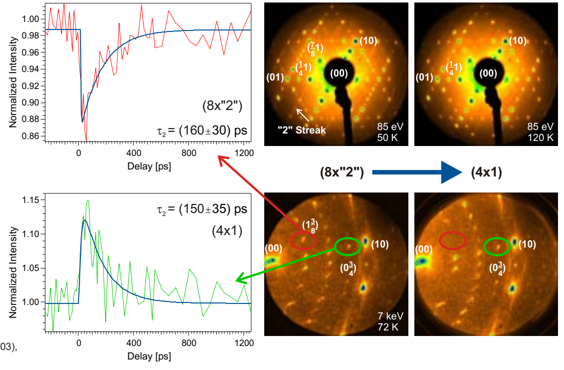


PEEM-image: D. Wall.

M. Hupalo et al., Surf.Sci. **493**, 526 (2001).

Indium (Phasetransition)

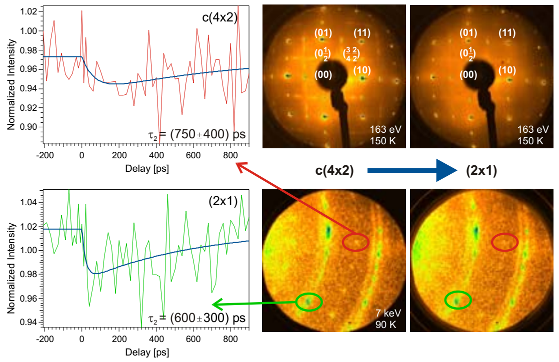
In monolayers were deposited on Si(111)-(7x7) at 700 K while monitoring with the RHEED.
In forms different reconstructions, the (4x1) has a coverage of 1ML. The (4x1) phase shows a phase-transition to (8x"2") at approximately 100 K.
This phasetransition is observed by preparing the (8x"2") and heating above the transition-temperature in a time resolved measurement.
The (8x"2")-diffraction spots disappear and the intensity of the (4x1)-spots increases.
The time constant τ_2 is in both cases 150 ps.



S. Mizuno et al., Phys. Rev. B **67**, 195410 (2003),
S. J. Rijkov et al., Surf. Sci. **588**, 15 (2001).

Clean Silicon

The order-disorder transition from Si(001)c(4x2) to (2x1) was investigated.
After flash annealing the sample at 1500 K a clear c(4x2) can be observed.
First preliminary results are shown here. The intensity drops by 4% upon excitation.
The intensity of the c(4x2) diffraction spots recover slowly with a time constant of several hundred ps.
These results agree surprisingly well with literature values on the excitation of the electron system.



T. Tabata et al., Surface Science **179**, L63 (1987),
M. Weinelt et al., Phys. Rev. Lett. **92**, 126801 (2004).