UNIVERSITÄT DUISBURG ESSEN

SFB 616

Ultrafast lattice heating in thin (semi-)metal films observed by time resolved electron diffraction

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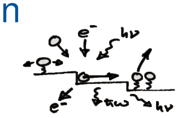


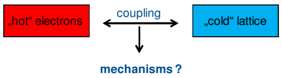
B3

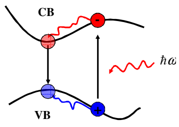
Introduction

The study of ultrafast (fs..ps) dynamics on atomic scales (several Å) requires ultrashort radiation pulses of sufficiently small wavelength or electron bunches witch kinetic energies of several tens of keV.
In our experiments, we use ultrafast electron diffraction (UED) of 30keV, sub-ps electron pulses from thin (semi-metal) films to observe the dynamics induced by femtosecond optical excitation. From the data obtained by our experiments, conclusions about the coupling between the – strongly heated – electronic system and the cold lattice system in different materials after this excitation can be drawn. The results are compared with the numerical solution of the two-temperature-model (TTM) and show excellent agreement with the results obtained by all-optical reflectivity and transmissivity measurements.

Motivation

- ultrafast (fs) optical excitation of solids
- relaxation and dissipation of electron energy due to
  - electron-electron-interaction (thermalization)
  - electron-phonon-coupling (lattice heating)

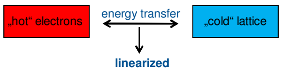




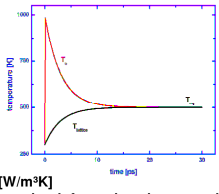
extreme nonequillibrium!

Two Temperature model (TTM)

- linearized coupling between electronic and phononic system



$$-c_e \frac{\partial T_e}{\partial t} = c_l \frac{\partial T_l}{\partial t} = G(T_e - T_l)$$



- G : material-specific electron-phonon-coupling constant [W/m³K]
- measure either transient electron or lattice temperature to gather information about coupling!

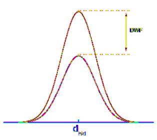
Debye-Waller-Effect

- decrease of diffraction peak intensity for „moving" lattice

$$DWF = \frac{I}{I_0} = e^{-\left(\frac{4\pi^2 <u^2>}{3\, d_{hkl}^2}\right)}$$

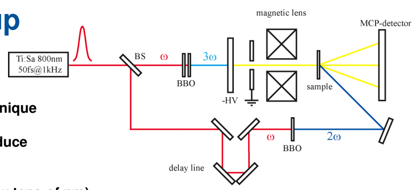
- Debye-approximation: $<u^2> \propto T_l / \Theta_D$

spot profile does not change



Diffraction Intensity depends on lattice temperature!

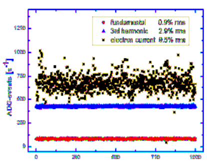
Experimental setup

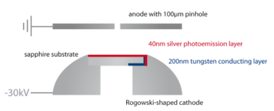


- optical pump - electron probe technique
- high-rep (1kHz) laser system to reduce space-charge-effects
- free-standing thin film samples (few tens of nm)

Electron source design and performance

- 30keV electrons – 0.07Å
- <1ps temporal resolution @ 10³ electrons/pulse
- 450 µm focal spot on MCP-detector
- 9.5% rms shot to shot stability (measured with channeltron detector)

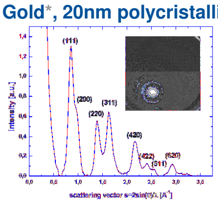


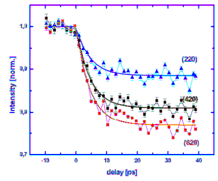


- 40nm silver photoemission layer for high electron yield [1]
- Rogowski-shaped photocathode for maximum field strength (10kV/mm)

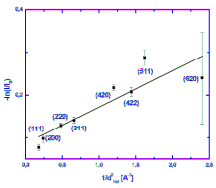
Time resolved measurements : Au and Cu

Gold*, 20nm polycristalline



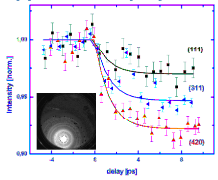


- excitation fluence 0.9mJ/cm²
- decrease depends on (hkl)
- time constant (6.4±0.5)ps
- „weak" coupling



relative decrease follows DWF!
slope yields <u²>

Copper*, 20nm polycristalline

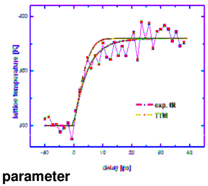


- 6.9mJ/cm²
- time constant (1.3±0.6)ps
- „strong" coupling

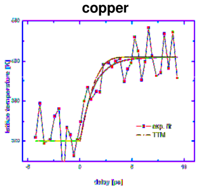
* samples provided by O. Posth and Ch. Hassel, AG Farle, University of Duisburg-Essen

Comparsion with TTM

gold

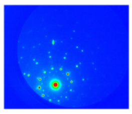


copper



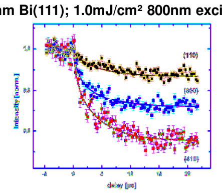
- numerically solve TTM with G as free parameter
- best fit : G=2.1·10^16 W/m³K (Au) and G=10·10^16 W/m³K
- excellent agreement with values obtained by all-optical methods [2,3]!

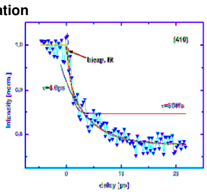
More complex relaxation behaviour in †Bi ?



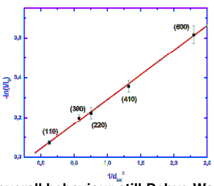
- epitaxially grown (111) high qualtity Bi films [4]
- 12 fold symmetry from two domains
- orders not sensitive to A1g phonon mode

22nm Bi(111); 1.0mJ/cm² 800nm excitation





biexponential drop!



overall behaviour still Debye-Waller ...

- two distinguishable decay channels for 15nm and 22nm films
- only monoexponential behaviour in thicker films (25, 30nm)
- possible indicator for thin film semimetal to semiconductor transition?

† made by Thomas Payer, AG Horn-von Hoegen University of Duisburg-Essen

Conclusion and Outlook

UED has been shown to be capable of measuring the energy transfer from hot electrons to lattice vibrations subsequent to femtosecond optical excitation of solids. The direct comparison with the TTM yields the corresponding coupling constants G. These values are useful in e.g. energetic ion bombardment of solids and optical ablation experiments as well as for the fundamental understanding of the interaction of short pulse radition with matter. Furthermore, a film thickness dependent relaxation behaviour in Bi has been observed, possibly indicating a strong dependence of the electronic structure from the sample thickness. However, further theoretical and experimental investigations are necessary for a complete picture of the dynamics induced by femtosecond optical excitation of these films.

[1] Aeschlimann et. al., „*A Picosecond electron Gun for surface analysis*", Rev. Sci. Instrum. **66** (2), (1995)
[2] Hohlfeld et al., „Electron and lattice dynamics following optical excitation of metals", Chem.Phys. **251** (2000)
[3] Elsayed-Ali et al., "Time-Resolved Observation of Electron-Phonon Relaxation in Copper", PRL **58**, 1212 (1987)
[4] Payer et. al.," Ultrathin Epitaxially Grown Bismuth (111) Membranes", Appl. Phys. Lett (accepted)
[5] Hoffmann et. al. "Semimetal-to-semiconductor transition in bismuth thin films", PRB **48**, 11431 (1993)

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