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

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Introduction
The study of ultrafast (fs-psec) dynamics on atomic scales (several Å) requires ultrashort radiation pulses of sufficiently small wavelength or electron bunches with kinetic energies of several ten 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)

Two Temperature model (TTM)
- linearized energy transfer between electronic and phononic system

Debye-Waller-Effect
- decrease of diffraction peak intensity for „moving“ lattice

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 – 10¹⁰ electrons/pulse
- 455 µm focal spot on MCP-detector
- 2.5% rms shot to shot stability (measured with channeltron detector)

Time resolved measurements: Au and Cu

More complex relaxation behaviour in Bi?
- 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?

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 radiation 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.

References

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