ESRF	**Experiment title:** Reducing thermal conductivity of tailored thin films for thermoelectric applications	**Experiment number**: MA2235
Beamline: ID09B	**Date of experiment**: from: 10-06-2014 14-06-2014	**Date of report**: 19-08-15
Shifts: 12	**Local contact(s)**: M. Wulff, D. Leshchev	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. A. Plech* (KIT Karlsruhe)

S. Eon*, Prof. H. Bracht (Uni Münster)

M. Daniel*, M. Albrecht (Uni Chemnitz)

Report:

The goal of the experiment was to deduce a change in the thermal conductivity in semiconductor structures that are coated by a multilayer stack of isotopically enriched sublayers of silicon or germanium. This arrangement is considered to provide a means of decoupling electronic conductivity from thermal conductivity in thermoelectric applications. A second set of samples consisted in Skutterudite layers, which, by introducing rattling atoms in the natural crystal cavities add phonon scattering centers that reduce thermal conductivity.

The approach in time-resolved X-ray scattering (TR-XRD), while already being established in analogy to optical spectroscopy (time domain thermal reflection [1]) and used successfully in earlier beamtimes [2,3], still holds some surprizes. In fact the thermal conductivity is being deduced by comparing the cooling behaviour of the multilayer stack of a covering gold film after laser excitation to that of a reference epilayer. By this approach different contribution to the thermal resistance can be decoupled (timewise and by comparison of the parallel grown reference layer). Indeed the interface conductance between gold and analysed layer stack is of high importance and needs to be quantified. Additionally a high resistance hinders the fast heat decay and thus the sensitivity on the transport in the structure below. Therefore a chromium layer aids bonding between gold and oxide and reduced the interface resistance [4]. Still we found one difference with theory that concerns the initial distribution and dissipation of heat. Two effects may add here: residual light transmission through the 25 nm gold film (at 800 nm) may directly deposit heat into the semiconductor and excited fast electrons deposit energy into the semiconductor as well.

When taking this effect into account a better fitting of the cooling curve is achieved on a longer delay scale and in principle the conductivity of the substrate below can be checked as well. After this careful analysis we still find a considerable reduction of thermal conductivity of the multilayer stacks as compared to the reference [5]. This observation is not explained by standard phonon band calculations and molecular dynamics simulations [6,7], but rather points towards additional mechanisms or contributions from disorder scattering. One yet neglected mechanism is indeed found, Umklapp scattering at the mini-zone edges of the multilayer stacks [8].

Concerning the skutterudite samples we still face the same complication, which is by far stronger, but still qualitatively we find the effect of increasing filling ratio of the guest atoms on a reduction of thermal conductivity.

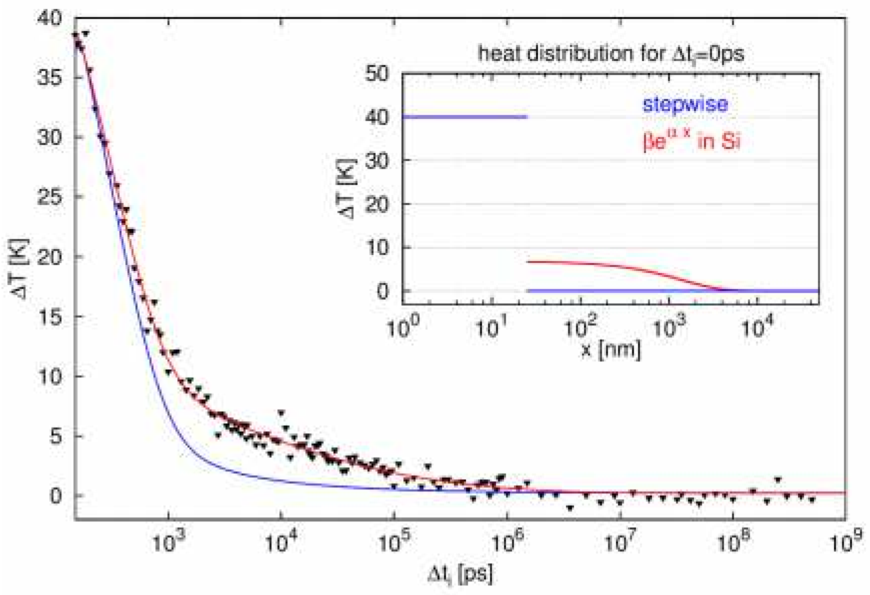

Fig. 1*: Temperature decay curves of gold films on top of the investigated silicon multilayer structure as determined from the time-resolved shift of the gold (111) powder peak after laser pulse excitation (triangles). Using a model with stepwise deposition of energy in the gold film and solution of the heat diffusion equations (blue lines) does not show a good fit, while an additional exponentially decaying profile within the silicon multilayer structure improves the model considerably (red lines). Initial temperature profile as inset.*

In summary TR-XRD for determination of thermal conductivity in surface-near regions is now well established, we have repeatedly found very strong reduction of conductivity with isotopic modulation in multilayer stacks and published the results in [2-5,8], accompanied by molecular dynamics simulations [6,7]. The mechanism is yet not clear and deserves further investigations.

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