ESRF	**Experiment title:** Heat Transfer in multilayers of pure silicon isotopes	**Experiment number**: SI 2242
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Report:

The goal of the experiment was to deduce a change in the thermal conductivity in silicon structures that are coated by a multilayer stack of isotopically enriched sublayers of silicon. This arrangement is considered to provide a means of decoupling electronic conductivity from thermal conductivity in thermoelectric applications. The used samples consisted in either a 10 nm ^{28}Si and 10 nm ^{29}Si stack of 20 repeat units or a natural silicon epilayer of 400 nm as reference. On top of these layers a thin film of metallic gold was deposited. The deposition involved either sputtering or deposition by evaporation, in the latter case with a thin chromium interlayer. The interlayer acted as a wetting layer for a better adhesion of the gold film.

The general idea is to hat the gold film with a 2 ps laser pulse while watching the cooling down via the peak shift of the gold powder scattering. The scattering (typically the (111) reflection of the preferentially oriented gold film) can serve as direct thermometer for the recording of the film temperature [1,2]. The cooling occurs via the substrate beneath. By means of pulsed x-ray scattering a time resolution of 100 ps is achieved, which is better than the typical cooling times of 1-5 ns.

By comparing the cooling times of the reference sample to the multilayer stack one can deduce the additional effect of the multilayer stack, which acts as a phonon barrier due to the mismatch of sound velocities.

The quality of the data was excellent and thanks to short exposure times per frame of fixed delay (2-5 secs) and a fast readout CCD (FRelon) we could record the cooling with fine steps over a long time span (typically 10 ns) for a series of samples. Before regarding the multilayer stack we already noted the strong dependence of the cooling times of the metal films for the different layer thicknesses and presence of chromium interlayer. This is indeed in agreement with theoretical considerations. If one considers the cooling of a thin film via substrate and only weak thermal coupling to it, then one would expect that the only parameters that enter the equation are film thickness (as heat reservoir) and interface conductivity that characterises the heat transfer from the film to the substrate. The heat flow only

depends on the difference in temperature across the interface, thus resulting in a exponential temperature decay with τ_K being the characteristic cooling time. In terms of formulae this is written as:

$$\tau_K = c\rho R_K d,$$

with c, ρ, being heat capacity and mass density of the layer, R_K and d the (inverse) interface conductivity and film thickness [3]. This is indeed what we have seen for the first 1-2 nanoseconds after pulse heating of the film. The decay of the temperature is exponential and a dependence on the film thickness is found. After some 2 nanoseconds, however, the cooling slows down, which is an indication that the substrate temperature can no longer be regarded as constant, as the heat conduction in silicon is finite.
Quantitative models for the full behaviour have been implemented for nanoparticles [2] and are also available for thin films [3, 4], which e are going to implement.
Moreover we found a (at first glance) rather surprising fact, that the chromium interlayered layers showed a much faster cooling time. At second thought this is quite natural, considering the better adhesion. A better contact of the film also increases the interface conductance.
In that framework an increase in cooling time due to a subsurface isotope multilayer has been seen in particular for the thinnest films (30 nm) and there quite clearly for the chromium samples. There the initial exponential decay is modified. We are currently modelling the full cooling curve in order to understand all effects.

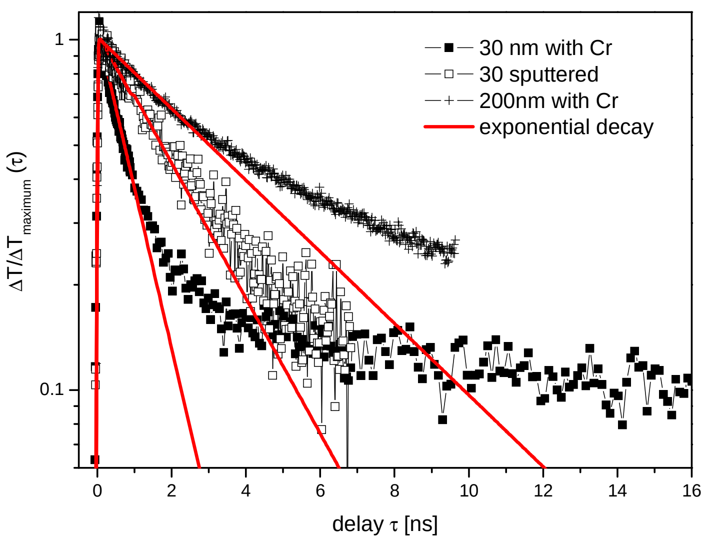

Fig. 1: *Normalized temperature decay of gold films of different thickness as function of delay τ after laser excitation. The lines are fits with an exponential decay.*

One conclusion out of these observations may already be noteworthy. At first glance one would tend to use only the data from very thin metal layers for deriving the modified interface conductance. However, considering that for the thicker films the bulk heat conductivity changes the exponential decay much stronger by long lasting tail in temperature after excitation; one can use this effect to gain depth information on the thermal properties of the material under investigation. A very thin metal layer only probes the interface and the very near subsurface volume, while the cooling of thicker films is also sensitive on the thermal properties in larger depths. We are still investigating the magnitude of the effect by simulations and ideally by additional experiments.

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[3] B. Krenzer, et al.: *Heat Transport in Nanoscale Heterosystems: A Numerical and Analytical Study*, J. of Nanomat. (2008) Article ID 590609, doi:10.1155/2008/590609.
[4] A. N. Smith, et al: *Thermal boundary resistance mesurements using a transient thermoreflectance technique*, Microscale Thermophys. Eng. 4 (2000) 51.