ESRF	**Experiment title:** Heat Transfer in multilayers of pure silicon isotopes	**Experiment number**: SI2530
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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.

Determining thermal conductivities in thin layers of surface-near regions of materials is not an easy task to perform. Instead of using optical techniques (time-domain thermoreflectance) we introduced time-resolved x-ray scattering as a direct probe for thin-film temperature [1,2]. The idea is to launch a heat wave in a material by heating a top layer by means of a short laser pulse. The decay of this heat wave into the bulk will be governed by the thermal conductivity in the surface-near region and thermal resistances at interfaces between dissimilar materials (the latter is often named Kapitza resistance). Adequate modelling of the surface-near structure with its conductivity profile allows gaining back the desired material parameters. This is in particular an issue in tailored thin film materials, such as the envisaged semiconductors.

In practical terms a thermal transducer film of gold is deposited on top of the investigated surface. This film can be excited by 400 nm laser pulses from the Ti:Sa femtosecond laser. Although the energy deposition in the gold film is shallower, an 800 nm laser beam has been proven to improve the stability of excitation. Stroboscopic x-ray pulses probe the lattice expansion of gold predominantly in direction perpendicular to the surface. This is facilitated by the preferential (111) oriented growth of the gold film, while in-plane directions can be probed in different geometry as well [2].

Heat flows into the material within a time scale of some nanoseconds, where the first time-limiting step is the crossing of the gold-substrate thermal barrier. It turned out that this interface resistance can be minimized by a chromium interlayer as adhesion agent [2]. The time scale additionally depends linearly on the gold film thickness. Best results were achieved with 30 nm gold films, but thinner films may give even shorter relaxation times [3]. Apart from this resistance heat will flow into the bulk according to the material thermal conductivity under investigation. Modelling of the heat transfer can be done by an analytical model in Laplace transform [2,4] or by numerical solution of the diffusion equations under inclusion of temperature drops wherever finite interface resistances are expected.

We have analysed different samples of silicon isotopes, i. e. alternating layers of 20x (10 nm + 10 nm) of alternating 28-Si and 29-Si, or 28-Si and 30-Si, as well as reference samples of natural silicon and pure (thick) layers of 28-Si. As for the counterpart of germanium, multilayers of natural Ge and 74-Ge were measured. The same materials of technical interest (doped samples) were also available. Fig. 1 shows a set of measurements on silicon multilayers, each compared to its reference sample. The lines are fits with optimized numerical solutions of the underlying heat diffusion equations.

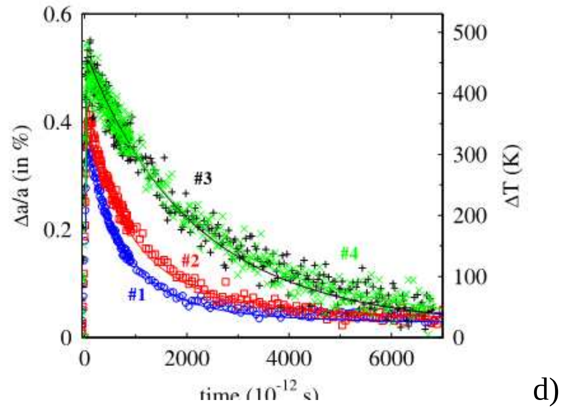
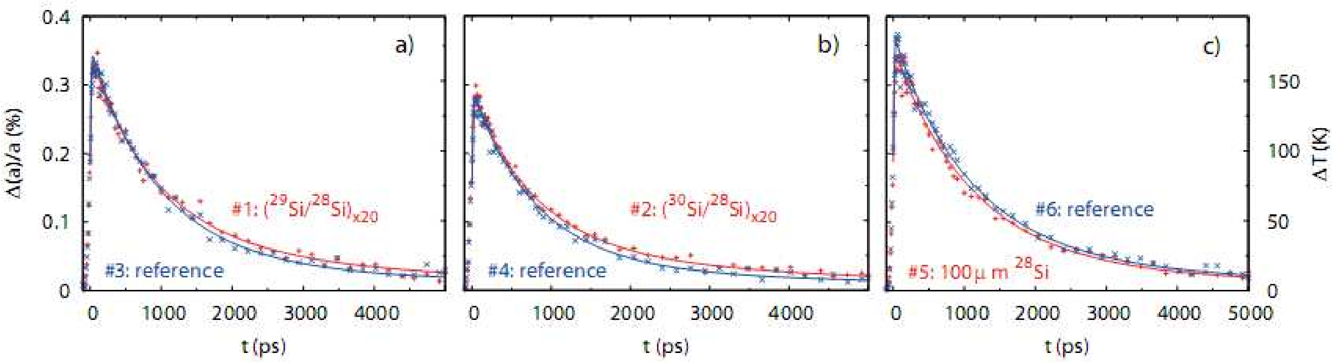


Fig. 1*: Temperature decay curves of gold films on top of the investigated silicon multilayers structures and reference substrates as function of delay time from laser heating. A)-B) multilayers with alternating isotopes 28 and 29, resp. 28 and 30. c) isotopically pure surface of 28-Si. D) Effect of gold film with (#1 reference and #2 multilayer) Cr interlayer and without (#3 reference and #4 multilayer).Conductivities are 196 W/mK for 28-Si, 150 for natural Si, and 50-60 W/mK for the multilayer structures.*



As a result there is a considerable reduction of the effective thermal conductivity of a silicon multilayer stack as compared to natural silicon. We could even resolve the 10 % effect of pure 28-Si versus natural silicon [5]. This demonstrates that a temperature resolution of down to 2 K is obtained, which allows discerning changes in conductivity of some percent. Furthermore the postulated reduction in conductivity of the multilayers from a simple acoustic mismatch model is even surpassed, pointing towards the coherent phonon transport problem in this multilayer stacks [5]. Similar conclusions are drawn from the germanium samples whilst having a lower density contrast. In conclusion a further optimization of the stacking sequence seems to be possible to further suppress heat conduction for thermoelectric applications.

[1] A. Plech, et al.: *Laser-Induced heating and melting of gold nanoparticles studied by time-resolved x-ray scattering*, Phys. Rev. B 70 (2004) 195423.
[2] D. Issenmann, S. Eon, N. Wehmeier, H. Bracht, G. Buth, S. Ibrahimkutty, A. Plech, *Determination of nanoscale heat conductivity by time-resolved x-ray scattering*, Thin Solid Films 541 (2013) 28.
[3] H. Bracht, N. Wehmeier, S. Eon, A. Plech, D. Issenmann, J. Lundsgaard Hansen, A. Nylandsted Larsen, J.W. Ager III, E.E. Haller, *Reduced thermal conductivity of isotopically modulated silicon multilayer structures*, Appl. Phys. Lett.101 (2012) 064103.
[4] A. N. Smith, et al: *Thermal boundary resistance measurements using a transient thermoreflectance technique*, Microscale Thermophys. Eng. 4 (2000) 51.
[5] H. Bracht, S. Eon, R. Frieling, A. Plech, D. Issenmann, D. Wolf, J. Lundsgaard Hansen, A. Nylandsted Larsen, J.W. Ager III, and E.E. Haller: *Thermal conductivity of isotopically controlled silicon nanostructures*, New Journal of Physics (2013), submitted.