ESRF	**Experiment title:** Umklapp process as thermal barrier in isotopically modulated multilayers	**Experiment number**: HC2539
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Report:

The goal of the experiment was to verify a change in thermal conduction in thin films of isotopically modulated semiconductor multilayers and connect the thermal diffuse scattering (TDS) to the excitation of Umklapp process that slow thermal conduction down. The samples consisted of multilayers of the silicon isotopes 28-Si and 30-Si and natural Ge and 70-Ge alternated by MBE growth in individual layers of 6 - 10 nm each [1]. When the multilayer is covered by a thin film of gold, this metal layer can be used as transducer to absorb laser pulses to heat up rapidly. Its temperature can be monitored to derive the cooling rate. The latter reflects the (cross plane) thermal conductivity of the material below. Isotopically modulated structures show a reduction in thermal conduction that can be beneficial in e.g. thermoelectrics.

Experimental setup:

The multilayer stack can be excited by picosecond laser pulses, while the X-ray beam probes the lattice expansion at a given delay to reconstruct the temperature history of the top transducer layer. In case of direct excitation lattice expansion and thermal diffuse scattering of the multilayer gives direct information on lattice excitation in the sample. The powder scattering of the gold film, respectively thermal diffuse scattering close to the (400) fundamental reflection of the multilayers is recorded on a CCD camera.

Data quality:

The beamtime was delivered a stable, pulsed beam. The divergence of the beam at the sample position proved a bit high to resolve the TDS close to the Bragg peak, but at a scattering vector difference of > 0.1 AA^-1 the discrimination was still sufficient.

Results:

Earlier experiments proved difficult to analyse as the thermal kinetics of the gold transducer layer did not well follow the predicted diffusion laws, even when including the Kapitza resistance between gold and silicon. Two possible sources of deviations were identified. (i) If the gold layer is too thin, a sputtered gold film will still show some pores that allow to leak light into the underlying layer structure, so that silicon is heated directly (see fig. 1, published in [2]). We could verify that this problem disappears for film thicker than about 40 nm, which were used in the following. We have faint indications that few fast electrons are able to escape from gold into silicon to cause an promt lattice expansion, but this effect is weak.

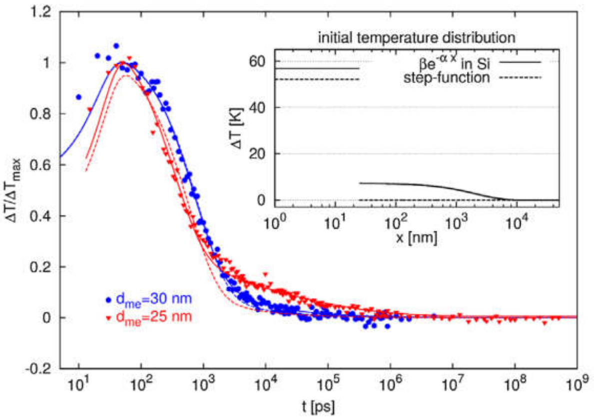

Fig. 1: *Temperature change of thin films of gold as function of delay t after laser excitation for different film thicknesses. For the thinnest film of 25 nm the temperature derives from the predicted cooling and has to be modeled by an initially heated sub-surface range of silicon (see inset) [2].*

(ii) Strong excitation (temperature rise in the gold film > 300 K) shows best signal-to-noise ratio in terms of temperature resolution, but at the same time risks of damaging the gold film. The high sensitivity on lattice expansion (> 2*10^-5) even detects subtle plastic deformation that adds a drift on expansion. This has been corrected for by recording signal at negative delay at each delay point of the measurement to discern plastic from elastic deformation.

With these improvements the modeling of cooling curves gave excellent fits to the data, allowing to extract subsurface thermal conductivities. Even the interface-related exponential cooling is discriminated from the bulk power-law cooling [3].

Finally, thermal diffuse scattering of silicon and germanium was recorded, which basically reproduced earlier findings of excess TDS within the mini-Brillouin zone of the multilayer, but did not resolve the zone edge further in relation to Umklapp processes. In fact, if Umklapp processes are considered to be incoherent, no coherent motion is to be seen. The cumulated data from several experiments was finally published in [3, 4].

Conclusions

The reduction of thermal conductivity in multilayered semiconductor structures, where the individual layers only differ by the nuclear mass of the isotopes has been verified for different structures and layer thicknesses and compared to models [3]. The behavior of the TDS in reciprocal space suggests a mechanism that is not governed by an incoherent acoustic impedance, but by Umklapp processes at the zone edge of the mini-Brillouin zone of the multilayer stack [4]

References:

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