From disorder towards tailored transport properties

Felix R. L. Lange[1,*)], K. Simon Siegert[1)], Ernst-Roland Sittner[1)] and Matthias Wuttig[1,2)]

[1)] I. Institute of Physics (IA), RWTH Aachen University, Sommerfeldstr. 14, 52074 Aachen,
[2)] JARA –Fundamentals of Future Information Technology, RWTH Aachen University, Germany
*lange@physik.rwth-aachen.de

ABSTRACT

Phase change materials (PCM) on the pseudo binary line between GeTe and Sb_2Te_3 (GST) are characterized by a rather unconventional high degree of disorder with randomly arranged vacancies on one lattice site. Here we present two ways to affect the degree of disorder in crystalline GST alloys and its effect on thermal and thermoelectric transport properties.

Key words: GST, disorder, thermal conductivity, thermoelectric transport properties

Phase change materials are a unique sub class of chalcogenides where both, an amorphous and a crystalline phase are stable at ambient conditions. This per se is not unique at all but the fact that, unlike in SiO_2 for instance, this phase transition can proceed on a nanosecond timescale [1]. This feature together with the pronounced property contrast between these two solid states renders PCM highly interesting for non-volatile data storage application.

One prominent class of PC alloys can be found on the pseudo binary line between GeTe and Sb_2Te_3. Compounds like $Ge_2Sb_2Te_5$ or $Ge_8Sb_2Te_{11}$ are already utilized in widespread rewritable optical data storage applications such as CD-RW or Blu-ray-RW, respectively. The peculiar point about these alloys is their high degree of disorder prevalent in the crystalline state that opens up a playground towards tailored transport properties. Upon crystallization the atoms form a meta-stable cubic phase where one site is occupied by Te and the second sublattice is randomly distributed by Ge, Sb and a stoichiometric amount of vacancies. Further heat treatment forces the cation site to reorganize each atom species into layers which finally triggers the transitions to the stable hexagonal phase. It is noteworthy to mention that this vacancy ordering appears to be a continuous process caused by annealing of the PC thin films.

Only recently Siegrist *et al.* reported a disorder induced insulator to metal transition in these compounds [2]. Depending on the annealing temperature the electrical conductance is either thermally activated or becomes metallic at elevated temperatures. While the carrier concentration stays almost constant the electrical resistance spans over six orders of magnitude at room temperature for different annealed PC films. Based on DFT calculations Zhang *et al.* identified clusters of vacancies to localize the wave functions responsible for charge transport in the early meta-stable cubic phase [3]. Dissolution of these clusters upon annealing then leads to a delocalization of the corresponding wave functions concomitant with the transition to the metallic state.

Of course disorder is not expected to solely affect electronic degrees of freedom. In fact we were able to demonstrate that the disorder prevalent in the early crystalline state of PCM can be so pronounced that the temperature dependence of the thermal conductivity κ resembles the one of a glass [4]. Furthermore we showed that annealing helps to enhance the thermal conductivity. By investigating various stoichiometries on the tie line (GeTe, $Ge_8Sb_2Te_{11}$, $Ge_3Sb_2Te_6$, $Ge_2Sb_2Te_5$ and $Ge_1Sb_2Te_4$) we were able to link this annealing effect to a reduction of point defect scattering that is concomitant with the ordering of the cation site.

Hence, with stoichiometry and annealing two pathways are revealed to control the degree of disorder in crystalline PCM. Interesting enough, the response in thermal and electrical transport properties on annealing differs for different stoichiometries (figure 1). This opens up a pathway towards independently tailored transport properties since, in principle, a wide range of (σ, κ) values can be accessed through appropriate stoichiometry and annealing conditions.

Using this concept we were only recently able to determine enhanced thermoelectric figure of merits ZT at elevated temperatures in $Ge_8Sb_2Te_{11}$ annealed to 250°C (figure 2) [5].

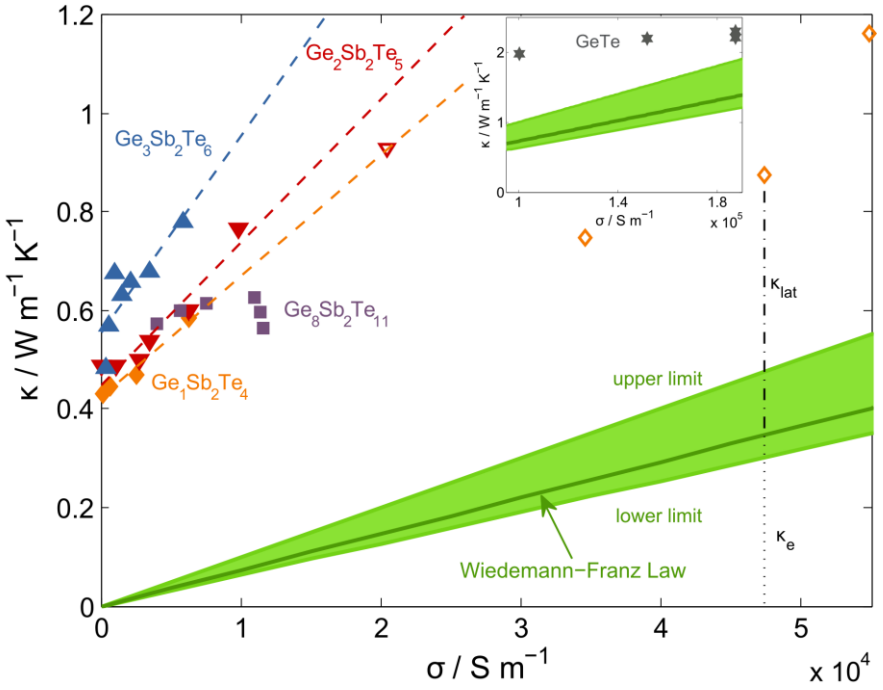

Figure 1: Thermal versus electrical conductivity for various Ge-Sb-Te alloys. Both, electronic and vibrational degrees of freedom are affected by annealing of the PC thin film. However, the response differs depending on the stoichiometry, which opens up a pathway towards independently tailored transport properties [4].

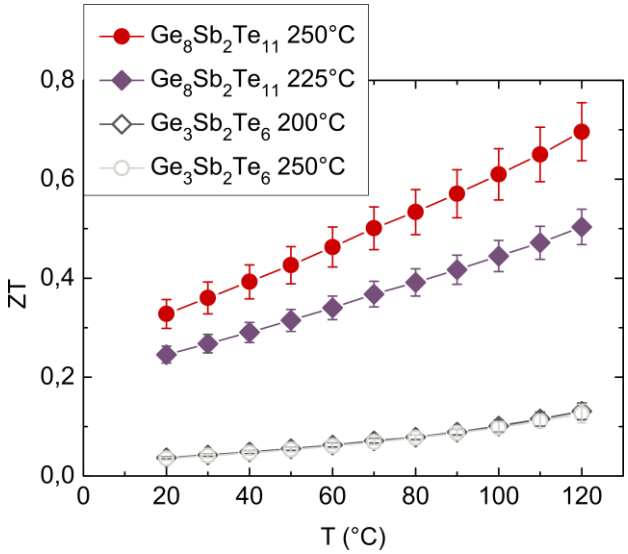

Figure 2: Thermoelectric efficiency as a function of temperature. The electrical properties in $Ge_8Sb_2Te_{11}$ can be tweaked without affecting the thermal conductivity (cf. figure 1) which results in enhanced thermoelectric figure of merits ZT at elevated temperatures [5].

REFERENCES

1. G. Bruns *et al.*, Appl. Phys. Lett. **95,** 043108 (2009)

2. T. Siegrist *et al.*, Nat. Mater. **10**, 202-208 (2011)

3. W. Zhang *et al.*, Nat. Mater. **11**, 952-956 (2012)

4. K. S. Siegert *et al.*, to be published

5. E.-R. Sittner *et al.*, Phys. Status Solidi A **210**, 147-152 (2013)

Biographies

Felix R. L. Lange obtained his diploma in physics at RWTH Aachen University in 2010. Under the supervision of Prof. Dr. M. Wuttig he worked for 12 months on the electrical properties of crystalline GeTe based pseudo-binary phase change materials. In February 2011 he started his Ph.D. at the I. Institute of Physics (IA) at the RWTH Aachen University expanding his investigations of highly disordered phase change alloys focusing on thermal and thermoelectric properties. Since July 2011 he is a doctoral researcher of the collaborative research center "SFB 917 Nanoswitches".

K. Simon Siegert has been working on phase change materials since 2008, when he started his diploma work at the I. Institute of Physics (IA) at the RWTH Aachen University under the supervision of Prof. Dr. M. Wuttig. During the course of his thesis, he focused on measuring the thermal conduction in PCM thin films by implementing the 3ω measurement technique. He received his diploma in 2009 and continued working at the I. Institute as a Ph.D. student. His current research concentrates on thermal transport properties of PCM as well as their thermoelectric potential.