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# Temperature-dependent thermal characterization of $\text{Ge}_2\text{Sb}_2\text{Te}_5$ and related interfaces by the photothermal radiometry technique

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**Abstract.** The thermal conductivity of  $\text{Ge}_2\text{Sb}_2\text{Te}_5$  (GST) layers, as well as the thermal boundary resistance at the interface between the GST and amorphous  $\text{SiO}_2$ , were measured using a PhotoThermal Radiometry experiment. The two phase-changes of the  $\text{Ge}_2\text{Sb}_2\text{Te}_5$  were retrieved, starting from the amorphous and sweeping to the *fcc* crystalline state at 130 °C and then to the *hcp* crystalline state at 310 °C. The thermal conductivity resulted to be constant in the amorphous phase, whereas it evolved between the two crystalline states. The thermal boundary resistance at the GST- $\text{SiO}_2$  interface was estimated to be higher for the *hcp* phase than for the amorphous and *fcc* ones.

## 1. Introduction

Phase change materials are extensively studied, due to their promising applications in the framework of Phase Change Memory (PCM) or Ovonic Unified Memory [1,2]. PCM functioning involves chalcogenide materials that are allotropic semi conducting elements and alloys belonging to the IV, V and VI group of the periodic classification. They can be reversibly brought from the amorphous to the crystalline state, so that the corresponding different electrical properties can be used for data storage.  $\text{Ge}_2\text{Sb}_2\text{Te}_5$ , commonly denoted GST, is one of the most popular chalcogenides [3], since it is stable at room temperature in the amorphous and hexagonal crystalline phases (*hcp*), and metastable in the face centered cubic phase (*fcc*) [4,5]. The transition temperature, is ~130°C for the *fcc*-crystalline phase and ~350°C for *hcp*-crystalline phase, whereas the melting temperature is approximately 600°C. The transformation between crystalline and amorphous phases is reversible: heating the amorphous GST to a temperature slightly above the glass-transition temperature leads to the crystalline phase; subsequent

heating to a temperature close to the melting temperature with fast quenching permits retrieving the amorphous phase.

The deposition of GST was achieved using DC-magnetron sputtering on a 100nm thick amorphous (a) SiO<sub>2</sub> thermally grown on Si substrate. In order to discriminate the GST thermal conductivity from the thermal boundary resistance (TBR)  $R_i$  at the GST-SiO<sub>2</sub> interface, 5 samples with different GST thicknesses, namely 100, 210, 420, 630 and 840 nm, were prepared. Amorphous SiO<sub>2</sub> layer thermal conductivity is  $k_{\text{SiO}_2} = 1.45 \text{ W m}^{-1} \text{ K}^{-1}$  and it does not vary significantly up to 400°C. The thermal resistance of the GST-SiO<sub>2</sub> stack is defined as:  $R = R_{\text{GST}} + R_i + R_{\text{SiO}_2} = R_t + R_{\text{SiO}_2}$ . Assuming that the Fourier law is valid in both the SiO<sub>2</sub> and GST layers, these two resistances can be expressed as  $R_{\text{SiO}_2} = e_{\text{SiO}_2} / k_{\text{SiO}_2}$  and  $R_{\text{GST}} = e_{\text{GST}} / k_{\text{GST}}$  for the SiO<sub>2</sub> and GST layers, respectively, where  $e_{\text{SiO}_2}$  and  $e_{\text{GST}}$  are the thicknesses of the layers. In this paper, we investigated the thermal conductivity of the GST layer, as well as the TBR at the interface between GST and amorphous SiO<sub>2</sub>, depending on temperature, from room temperature (RT) up to 400°C. Starting from the amorphous state, the GST was swept to the *fcc* and *hcp* crystalline states by increasing the temperature.

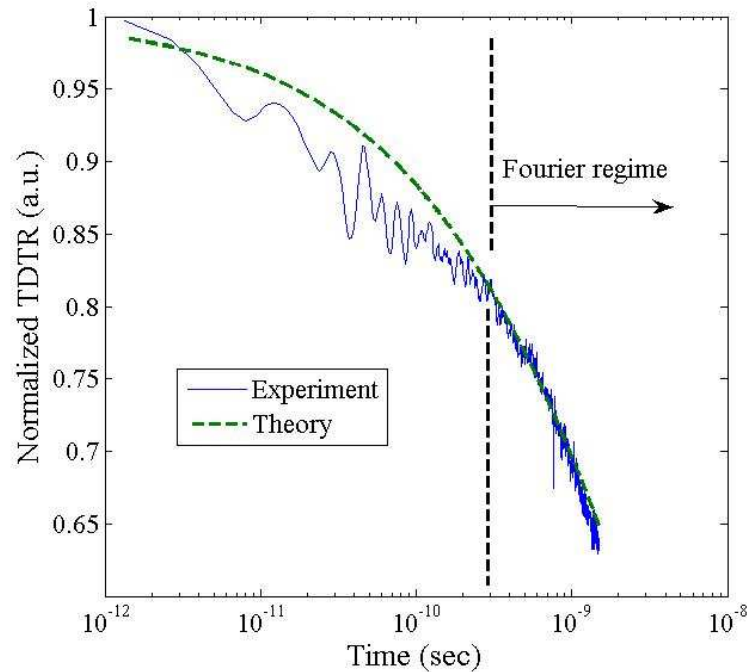
## 2. Experimental procedure

PhotoThermal Radiometry (PTR) experiments were implemented to measure the thermal resistance  $R_t$  of each sample, as a function of temperature. A platinum layer (30 nm thick) was deposited by e-beam evaporation on the GST layer as a transducer for the incident laser beam. In order to perform experiments at different temperatures, the temperature of the sample was controlled by a commercially available heating device, working in inert atmosphere (Ar). The sample was heated at a rate of 20 °C/min and annealed for 5 min at the required temperature before starting the measurement. The thermal excitation was generated on the sample surface by an Ar<sup>+</sup> laser of 514 nm wavelength and 1.7 W maximum power. The laser was modulated by an acousto-optic modulator using the square signal issued from a function generator and was reflected to the sample surface by a set of mirrors. The laser beam had a Gaussian profile of power repartition on the spot of 1 mm in diameter at  $1/e^2$ . A very fast photodiode was used to measure the reference signal, in order to avoid the phase lag due to the acousto-optic modulator driver. The thermal response was measured by an infrared HgCdTe detector. The wavelength measurement range of this IR detector was comprised between 2 and 13 μm. Parabolic mirrors coated with high reflective rhodium (reflectivity of 98% in the infrared detector wavelength band) were used to collect the emitted infrared radiation and to focus it on the infrared detector. The detector wavelength operating range was higher than that of the laser, therefore the measurement was not disturbed by the photonic source; moreover an optical filter was used in order to reject all the visible radiation arriving on the IR detector. The zone viewed by the detector was the image of the infrared sensitive element on the sample corresponding to a circle of 1 mm in diameter. A lock-in amplifier was used to measure the amplitude and the phase lag between the reference and the detector output, according to the frequency. The frequency range swept during the experiment was [1-100] kHz. Each measurement for the amplitude and phase was affected by a 5% standard deviation. The periodic temperature variation  $\Delta T$  at the sample surface was small enough to assume that the measured radiative emission by the IR detector was linearly proportional to  $\Delta T$ .

## 3. Validation of Fourier's law application

The goal of this section is to justify the expression of the thermal resistance for each layer (GST and  $\alpha$ -SiO<sub>2</sub>) according to their respective thermal conductivities  $k$  ( $R = e/k$ ). We first performed a picoseconds time domain thermoreflectance (TDTR) experiment on the 400 nm thick GST layer in the *hcp*-crystalline phase at room temperature. The experimental method is described in the paper of Battaglia et al. [6] and is based on a time resolved pump-probe setup, using ultra short laser pulses (wavelength = 500 nm and pulse duration  $\tau = 100$  fs) generated by a Ti:sapphire laser. The transducer is

an aluminum film (denoted Al, with:  $e_{Al} = 55\text{ nm}$  for the thickness,  $\rho C_p(\text{Al}) = 2700 \times 900\text{ J kg}^{-1}\text{ m}^{-3}$  of specific heat per volume unit), deposited on the GST layer in order to increase the signal-noise ratio during the TDTR. The expression of the average (with respect to the spatial distribution of the temperature on the heated area) normalized time domain thermoreflectance signal is:  $\overline{\text{TDTR}} = \exp(\alpha^2 t) \text{erfc}(\alpha \sqrt{t})$ , where  $\alpha = E_{\text{GST}} \tilde{\beta}_h / \rho C_p(\text{Al})$ ,  $E_{\text{GST}} = \sqrt{k_{\text{GST}} \rho C_p(\text{GST})}$  is the effusivity of the GST layer and  $1/\tilde{\beta}_h = e_{Al}$  is the heat penetration depth during the thermalization process between electrons and the lattice in the aluminum film. The result of the experiment is reported in the figure 1, as well as the simulation obtained from the model. It clearly appears that the measured impulse response fits very well with the semi-infinite behavior when time becomes higher than  $\Delta\tau_{\text{min}} = 0.3\text{ nsec}$ . This demonstrates that the Fourier law can be used for GST layers whose thickness is higher than  $\min(e_{\text{GST}}) \approx \sqrt{\Delta\tau_{\text{min}} k_{\text{hcp-GST}} / \rho C_p(\text{GST})} = 19.3\text{ nm}$  (for this calculus we used  $k_{\text{hcp-GST}} = 1.7\text{ W m}^{-1}\text{ K}^{-1}$  and  $\rho C_p(\text{hcp-GST}) = 6400 \times 212\text{ J m}^{-3}\text{ K}^{-1}$ ). This value can be viewed as the phonons mean free path in the hcp-GST and it must be lower in the amorphous phase. Implicitly, it demonstrates also the validity framework of the thermophysical properties of GST in the hcp-phase. The same experiment was performed on a  $\alpha\text{-SiO}_2$  layer and it was found that  $\min(e_{\alpha\text{-SiO}_2}) \approx 5.3\text{ nm}$ .

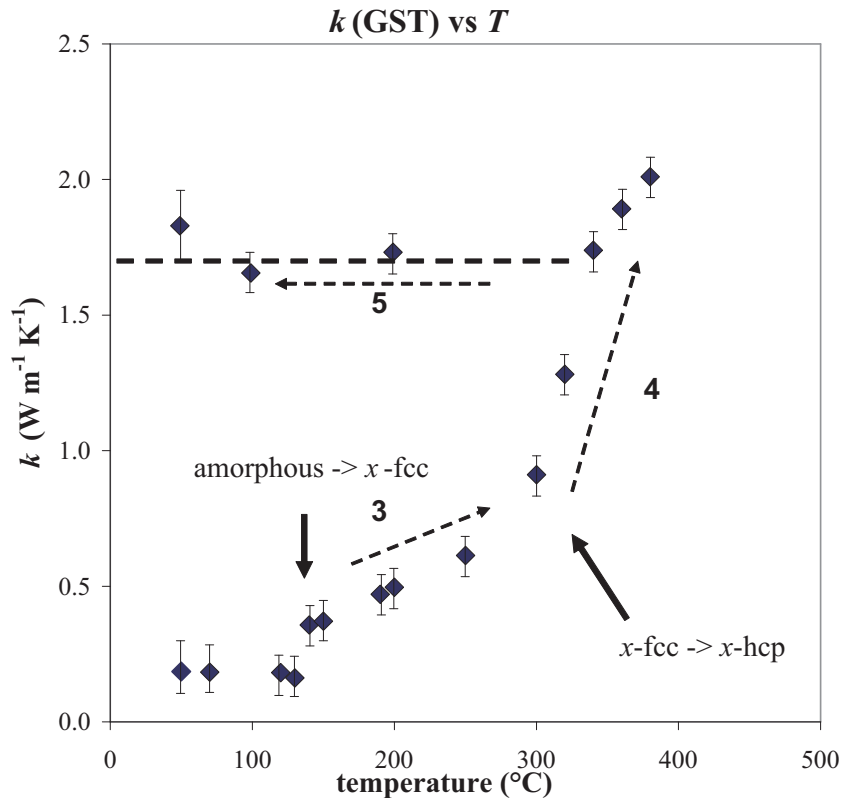


**Figure 1.** Measured impulse response using the TDTR.

#### 4. Results

Using the PTR it is found that the intrinsic thermal conductivity for the *fcc*-GST varied from  $k_{\text{fcc-GST}} = 0.42\text{ W m}^{-1}\text{ K}^{-1}$  to  $0.91\text{ W m}^{-1}\text{ K}^{-1}$  in the  $140^\circ\text{C}$ - $300^\circ\text{C}$  range (see figure 2). From the phase change temperature to  $250^\circ\text{C}$ , the TBR decreased significantly, to reach a minimum value of  $R_i = 5 \times 10^{-9}\text{ K m}^2\text{ W}^{-1}$ . Above  $250^\circ$ , the TBR increased to  $9 \times 10^{-8}\text{ K m}^2\text{ W}^{-1}$  at  $400^\circ\text{C}$ . The intrinsic thermal conductivity for *hcp*- GST (from  $310$  to  $400^\circ\text{C}$ ) varied from  $k_{\text{hcp-GST}} = 1.1\text{ W m}^{-1}\text{ K}^{-1}$  to

$2 \text{ W m}^{-1} \text{ K}^{-1}$ . Obviously, this particular behavior can not be observed when one characterizes an annealed sample at room temperature. Indeed, the thermal conductivity of the *hcp*-phase remained quite constant and equal to  $1.6 \text{ W m}^{-1} \text{ K}^{-1}$ . On the other hand, it was found that the TBR between the GST and  $\alpha\text{-SiO}_2$  is higher ( $R_i \approx 1 \times 10^{-7} \text{ K m}^2 \text{ W}^{-1}$ ) for *hcp*-GST than for *fcc*-GST and  $\alpha$ -GST.



**Figure 2.** Measured thermal conductivity using the PTR.

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