Experimental report

27/03/2020

Proposal: 6-05-984 **Council:** 10/2016

Title: Study of the phase separation in GexTe100-x amorphous films

Research area: Materials

This proposal is a new proposal

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Samples: GexTe100-x with x=12, 20, 27, 30, 37, 42 and 48 at.% Ge

Instrument	Requested days	Allocated days	From	To
D11	4	1	20/02/2017	21/02/2017
D16	4	0		

Abstract:

Storage density is a key-issue in the field of information storage. Storage densities in PC-RAM memories could be drastically increased thanks to phase-change materials that could offer the possibility of making multi-level cells. Changing its amorphized volume could systematically alter the cell resistance. This process requires, however, a stable amorphous state, i.e., a material with a time-independent resistance. This effect, commonly denoted as resistance drift, may cause severe data corruption over time and thus hampers the realization of multi-level phase change memories.
We recently combined the amorphous state resistivity measurements to structural and thermal analyses in amorphous GexTe100-x films. Our results allow us to identify two singularities, which could be linked to a phase separation in films with $x > 25$ or $x > 35$ at.% Ge. In this context, we propose to extend the structural investigations of the Ge-Te system by exploring possible structural rearrangement using small angle neutron scattering (SANS). These measurements will help to understand the ageing of the Ge-Te films, which could be interesting in the operation mode of electrical memories.

Study of the phase separation in Ge_xTe_{100-x} amorphous films

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Abstract

Storage density is a key-issue in the field of information storage. Storage densities in PC-RAM memories could be drastically increased thanks to phase-change materials that could offer the possibility of making multi-level cells. Changing its amorphized volume could systematically alter the cell resistance. This process requires, however, a stable amorphous state, i.e., a material with a time-independent resistance. This effect, commonly denoted as resistance drift, may cause severe data corruption over time and thus hampers the realization of multi-level phase change memories.

We recently combined the amorphous state resistivity measurements to structural and thermal analyses in amorphous Ge_xTe_{100-x} films. Our results allow us to identify two singularities, which could be linked to a phase separation in films with x > 35 at.% Ge.

In this context, we propose to extend the structural investigations of the Ge-Te system by exploring possible structural rearrangement using small angle neutron scattering (SANS). These measurements will help to understand the ageing of the Ge-Te films, which could be interesting in the operation mode of electrical memories.

Experimental Details

Three glasses of compositions $Ge_{15}Te_{85}$, $Ge_{24}Te_{76}$ and $Ge_{40}Te_{60}$ were prepared from 5N (99.999% purity and metallic impurities <200 ppm) germanium chips (Aldrich) and 4N5 (99.995% purity) tellurium powders (5NPlus). Different elaboration techniques, as twin roller quenching (for $Ge_{15}Te_{85}$ and $Ge_{24}Te_{76}$ glasses) and co-thermal evaporation (for $Ge_{40}Te_{60}$ glass), were used depending of the composition (for more details of sample preparation refer to Refs. [1, 2].

Small Angle Neutron Scattering (SANS) was carried out at D11 instrument at the Institute Laue-Langevin in Grenoble (France). The wavelength was set to 4.6982 Å and different instrument configurations were used to cover a Q range from 2.10^{-3} to 0.5 $Å^{-1}$ (sample-detector distances: 1.6 m, 8 m, 16 m, 20 m and 39 m). The raw scattering data were corrected for the electronic background and empty cell and were normalized on the absolute scale in cm^{-1} using the attenuated direct beam to calculate the incident flux and using the ILL Lamp software for data reduction. Powder samples obtained by twin roller quenching or co-thermal evaporation were placed in aluminum envelopes. Measurements were carried out at room temperature.

Results

Figures 1 shows normalised SANS scattering intensities I(Q) for the three samples of the Ge_xTe_{100-x} system measured at room temperature. All curves exhibit a change from a flat regime at high-Q values to a regime with a linear increase (in log-log) at low-Q values. However, some differences can be observed when the germanium content is increasing in the glass. We observe that the curves corresponding to $Ge_{15}Te_{85}$ and $Ge_{24}Te_{76}$ are practically superimposed, while $Ge_{40}Te_{60}$ sample shows differences particularly in the range of 0.01 to 0.2 $Å^{-1}$.

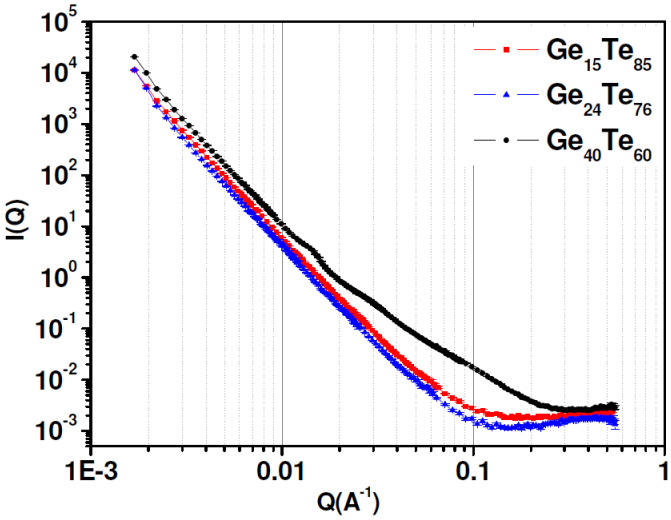

***Figure 1**: Normalised SANS scattering intensity I(Q) for Ge_xTe_{100-x} glasses elaborated by twin roller quenching or co-thermal evaporation technique.*

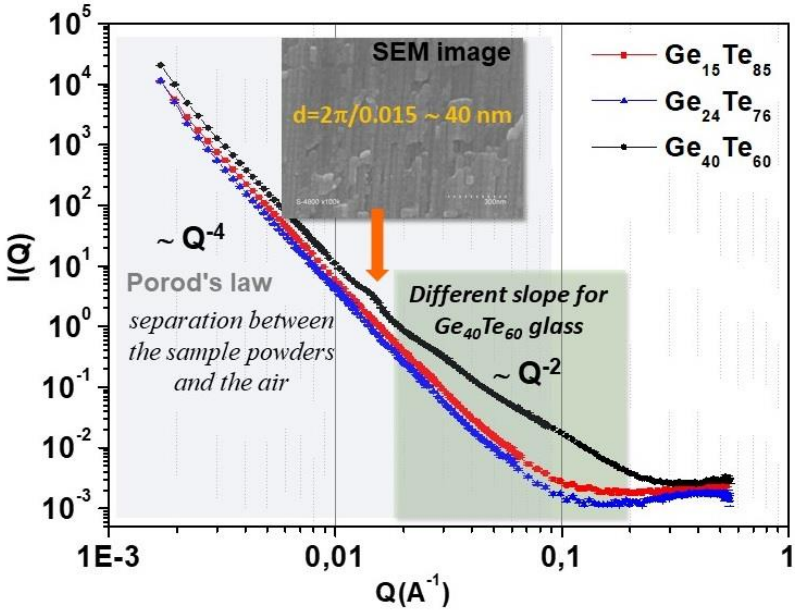

***Figure 2**: Normalised SANS scattering intensity I(Q) for Ge_xTe_{100-x} glasses. Different Q^{-m} regimens are shown. The SEM image show the topography of the $Ge_{40}Te_{60}$ thick film.*

The enlarged Porod's law, $I(Q) \approx BQ^{-m}$, could allow gaining information on the interface between the two media (see Figure 2).

- For $Ge_{15}Te_{85}$ and $Ge_{24}Te_{76}$ glasses a single regime is observed in the Q range [2.10^{-3} $Å^{-1}$,~ 0.1 $Å^{-1}$]. A similar slope very close to − 4 is obtained for both glasses. This corresponds to a Porod's law ($I(Q) \approx S.Q^{-4}$) characteristic of a two-phase sample with well-defined phase boundaries, which corresponds in our case to the abrupt separation boundaries between the sample powders and the air.
- For $Ge_{40}Te_{60}$ glass two different power law regimes are observed: a first regime at low-Q range [2.10^{-3} $Å^{-1}$, ~ 0.015 $Å^{-1}$] and a second regime at intermediate Q range [~ 0.015 $Å^{-1}$, ~ 0.1 $Å^{-1}$]. Whereas the slope is practically constant and close to m = 4 in the low-Q range, a particularly interesting behaviour is observed at intermediate Q range. In this Q-region, m is close to 2, which could be related to a lamellar phase separation. Moreover, this sample presents a repeating distance of about 40 nm which could correspond to the topography of the film (as it can be observed in the SEM image).

These results could confirm a phase separation for the Ge-rich sample, *i.e*. $Ge_{40}Te_{60}$, in agreement with the proposition of ref. [3], however it is important to note that the elaboration technique is not the same for all samples.

These results are very preliminary, and new experiments will be performed at the D16 instrument (Proposal 6-05-990) in order to confirm these interesting behaviours on a larger number of compositions that will all be elaborated by the co-thermal evaporation technique.

References

[1] A.A. Piarristeguy, E. Barthélémy, M. Krbal, J. Frayret, C. Vigreux, A. Pradel; « *Glass formation in the Ge_xTe_{100-x} binary system: synthesis by twin roller quenching and co-thermal evaporation techniques* », Journal of Non-Crystalline Solids 355 (2009) 2088-2091.

[2] P. Jóvári, A.A. Piarristeguy, R. Escalier, I. Kaban, J. Bednarcik, A. Pradel; « *Short range order and stability of amorphous Ge_xTe_{100-x} alloys ($12 \leq x \leq 44$)* », Journal of Physics: Condensed Matter 25 (2013) 195401.

[3] A.A. Piarristeguy, M. Micoulaut, R. Escalier, P. Jóvári, I. Kaban, J. van Eijk, J. Luckas, S. Ravindren, P. Boolchand, A. Pradel; « *Structural singularities in Ge_xTe_{100-x} films* », Journal of Chemical Physics 143 (2015) 074502.