

# Aging mechanisms in amorphous GeTe

Jean Yves Raty<sup>(1)</sup>, Wei Zhang<sup>(2)</sup>, Jennifer Luckas<sup>(3)</sup>, Riccardo Mazzarello<sup>(2,4)</sup>, Christophe Bichara<sup>(5)</sup> and Matthias Wuttig<sup>(3,4)</sup>

(1) Physique de la Matière Condensée, B5, Université de Liège, B4000, Sart Tilman, Belgium

(2) Institute for Theoretical Solid State Physics, RWTH Aachen University, 52056 Aachen, Germany

(3) I. Institute of Physics (IA), RWTH Aachen University, 52056 Aachen, Germany

(4) JARA Fundamentals of Future Information Technology, RWTH Aachen University

(5) CINaM, CNRS and Aix-Marseille University, Campus de Luminy, Case 913, F13288, Marseille, France

## ABSTRACT

We investigate the structure of amorphous GeTe using Density Functional Theory based Molecular Dynamics, using either the standard Generalized Gradient Approximation, or the more elaborate van der Waals approximation that proves more accurate in this system. New insight is provided on the stability of homopolar GeGe bonds and tetrahedral Ge bonding, in relation with the resistance drift phenomenon, that is investigated experimentally using photothermal deflection spectroscopy experiments.

**Key words:** GeTe, aging, resistance drift, Molecular Dynamics, DFT, photothermal deflection spectroscopy.

## 1. INTRODUCTION

Aging phenomena are common to all amorphous structures, but of special importance in phase change materials (PCM) since it impedes the realization of multi-level memories. Different interpretations have been proposed, but we focus here on the structural relaxation of amorphous GeTe, chosen because it is the simplest system that is representative of the wider class of GST alloys, lying along the GeTe-Sb<sub>2</sub>Te<sub>3</sub> composition line of the GeSbTe phase diagram.

## 2. EXPERIMENTS

Since the structural relaxations concerned with the drift take place on long time scales, the task of understanding them to limit their consequences is not a simple one. We successfully achieved this goal by developing new methods to overcome a series of hurdles. The issues we addressed and the solutions we developed are listed below.

- a) Directly generating an amorphous structure by quenching a liquid using Density Functional Theory (DFT) based Molecular Dynamics leads to one sample with a small number of atom (typically a few hundreds), and, hence of small number of atomic

environments. Here we sample a large number of local atomic environments, corresponding to different bonding schemes, by chemically substituting different alloys, selected to favor different local atomic structures. This enables spanning a larger fraction of the configuration space relevant to aging.

- b) GST alloys are known to display complex bonding mechanisms, for which the simple chemist's "octet-rule" does not apply, leading a long series of controversies, concerning in particular, the local structure around Ge atoms. We overcome this problem by using state of the art non local DFT-MD, including the so-called van der Waals corrections. This leads to more clearly defined environments that are then thoroughly analyzed.
- c) To extract some unifying picture of the amorphous structure we calculate local energies using a new approximate method that enables us to identify the driving forces for the structural relaxation.

The amorphous structures generated agree with all known experimental data, and the calculated electronic properties nicely match the most recent photothermal deflection spectroscopy experiments that are presented here.

#### **4. CONCLUSION**

Our results support a model of the amorphous phase and its time evolution that involves an evolution of the local (chemical) order towards that of the crystal (by getting rid of homopolar bonds), and an evolution of its electronic properties that drift away from those of the crystal, driven by an increase of the Peierls-like distortion of the local environments in the amorphous, as compared to the crystal.