Experimental and *ab-initio* investigations of sub-ps optical excitation effects in amorphous GeTe thin films

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ABSTRACT

Chalcogenide Phase-Change Materials (PCMs), mainly GeSbTe-based alloys, have already been widely used for optical data storage in DVD-RAM or CD-RW. Thanks to their unique reversible and very fast amorphous to crystalline phase transition, which is characterized by an uncommon huge change in optical and electrical properties, PCMs are now extensively studied. The aim is to develop innovative emerging non-volatile memories such as phase -change random access memory (PCRAM) in order to replace current dominant Flash memory technology or storage class memories (SCM) aiming at bridging the gap in between DRAM and NAND Flash technologies[1]. The interaction of PCMs with a fs light pulse has attracted significant attention due to fundamental interest since the possible non-thermal amorphous↔crystal phase transition could be used as a process to drive the phase-change above the thermal "speed limits"[2]. Our work addresses the investigation of the effect of high electronic excitation induced by a fs laser pulse on the femtosecond timescale in amorphous GeTe thin films. Frequency domain interferometry is used to measure the dielectric constant of the excited material during the first 9 ps after the excitation. Experimental results are compared to *ab initio* simulations performed in the specific non-thermal equilibrium conditions, i.e. the temperatures of the electrons and atoms being different. The aim is to get a fundamental understanding of the main mechanisms that drives the phase-change with the perspective to ultimately being able to push further its limits.

Key words: femtosecond laser, amorphous GeTe, non-thermal phase transition.

1. INTRODUCTION

This study targets to understand what happen to the GeTe material after first instants following the optical excitation used to induce a phase transition. Our experimental method is based on a pump-probe experimental setup: a first pump laser pulse excites the sample while a delayed pulse is probing the excited sample. Using femtosecond lasers allows us to monitor any evolution of the sample with a 100 fs resolution during the first 9 ps.

2. TWO COLORS POLARIZATION RESOLVED FREQUENCY RESOLVED FREQUENCY DOMAIN INTERFEROMETRY

Frequency domain interferometry (FDI)[3] is a pump-probe experiment that gives access to the variation of the refractive index of a material (**Fig. 1**). A pump pulse (25 fs, 800 nm, 1kHz) is used to trigger a phase transition. The probe beam is made of two pulses (120 fs, 532 nm) delayed by 9 ps in our case which are focused on the pump/sample interaction point. The first probe pulse impinges the surface of the sample before the pump pulse, and is thus reflected on the unperturbed material, whiles the second one that arrives after the pump pulse, is reflected on the pump-heated material. Both pulses are then sent in a spectrometer where they interfere in the frequency domain. The intensity variation and phase shifts in the interference pattern can be used to retrieve variations of the optical constant of the heated material. The interference pattern is simultaneously measured for the S and P polarization independently.

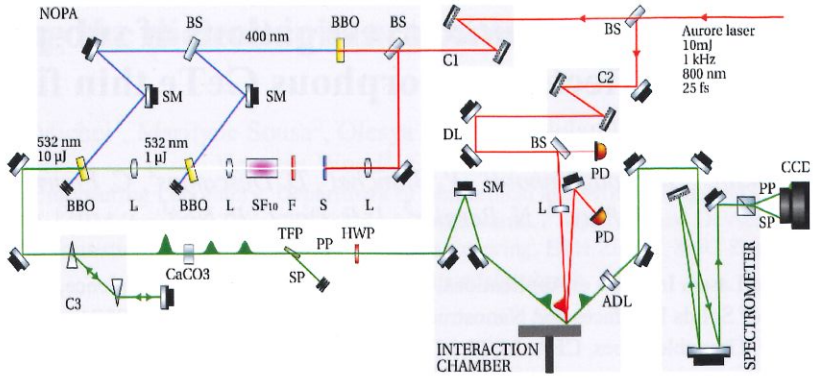

Figure 1: Schematic description of the FDI experimental setup.

3. RESULTS & DISCUSSION

We will present the results obtained on prototypical 500 nm thick amorphous GeTe thin films. Experiments have been conducted in the pump fluence range from 16 to 80 mJ/cm^2, allowing us to excite the amorphous material high enough to trigger a phase transition. Dynamics on the sub-ps time scale shows a very rapid transition evidenced on the real part of the refractive index. The polarization resolved FDI allows also us to foster information on the behavior of the surface which shows no formation of plasma: the sample is still solid or liquid. Moreover clear negative phase shift is attributed to a contraction of this surface, in the nm range after 2 ps. In order to grasp further understanding of the experimental results, *ab initio* simulations were performed for different electronic temperature up to nearly 1eV while keeping the atoms at 300K. The excited structures obtained are used to calculate the dielectric constants. A good qualitative agreement with experimental data is obtained. Changes induced by the electronic excitation will then be discussed in details.

4. CONCLUSION

Our experiments coupled to *ab initio* simulations evidence a change of local ordering triggered by the electronic excitation. Upon laser excitation, the pristine semi-conductive amorphous GeTe film experiences a clear change of its electronic Density of State and thus its electronic properties resulting from clear changes in its local amorphous structure. This result will shed new light on the fundamental understanding of processes of phase transition in phase-change materials.

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Biographies

P. Martinez obtained a research Master in Fundamental Physics, specialized in the study of 'Laser, matter and nanosciences' in 2018 in the University of Bordeaux. In October 2018, she started her PhD thesis on the "Ultrafast dynamics of phase change materials" at the CEntre of Intense Lasers and Applications (CNRS- UBordeaux, CEA). The main aspects of this study are to solve the dynamics of phase transitions coupling femtosecond time-resolved experiments and *ab initio* simulations thanks to a detailed parametric study of the excitation conditions. This work is done in close collaboration with P. Noé (CEA-LETI Grenoble – chalcogenide science and materials) and J.-Y.Raty (ULiège, CEA-LETI – theory and *ab initio* simulations).