Time Resolved Measurements of the Optical Recording Process at the Te Edge

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Sub-nanosecond time-resolved micro x-ray absorption measurements have been used to probe dynamical changes in the local structure about Te atoms in the phase change alloy Ge₂Sb₂Te₅ during the optical recording process using a optical pump/x-ray probe technique. Our experimental results suggest that the highest temperature state may not be molten during the optical amorphization process, but in fact may represent an excited state implying that at least for shorter laser pulses (500ps), there may be a non-thermal, electronic contribution the transformation. Keywords: Ge₂Sb₂Te₅, XANES, pump-probe experiment

Introduction: Recent developments in multimedia applications as well as the nature of the internet have lead to massive and ever increasing storage needs. One of the most attractive approaches to achieve this both from archival storage and from online usage perspectives is optical storage. In re-writeable optical media, data in encoded by taking advantage of the large changes in optical properties between the crystalline and amorphous phases of so-called phase change material of which Ge₂Sb₂Te₅(GST) is the prototypical alloy.

Based in large part upon observations of local structure of both the crystalline and re-amorphized phases of various Ge-Sb-Te alloys, our research group at AIST has developed an atomistic model of the switching mechanism for Ge-Sb-Te alloys. In this model, the large changes in electronic/optical properties are associated with a switch from a resonant-like bonding structure in the crystalline phase to a classical 8-N coordination in a glass like state [1]. In nucleation limited materials such as GST, atomistic simulations show that local Ge coordination with Te atoms plays a critical role. The gigapascal order transient pressures induced in the recording process as well as the presence of strong optical excitation makes it difficult to reproduce switching conditions in static measurements. Dynamic measurements hold the promise of leading to deeper knowledge of the changes in local order and potentially offer insight into novel techniques to optimize the



Fig. 1. A schematic representation of the experimental setup for the pump-probe experiment

tradeoff between storage lifetime (activation energy for transition between phases) and switching speed.

Experiment: A schematic of the experimental setup is show in figure 1. The sample was fabricated upon a 6 cm diameter fused quartz disk with the structure: substrate, a 50 nm thick GST layer, a 35 nm thick ZnS-SiO₂ layer, capped with a 1 micron thick Al-Cr alloy. An approximately 20 micron diameter optical pump beam with wavelength 532 nm and of 500 ps duration was directed through the transparent substrate towards the GST laver. An approximately two micron x-ray beam was directed coaxially with the pump beam from the metallic side of the sample. The detector was place perpendicular to the incident beam; the detector was gated to measure only x-rays from the relevant bunch. Using a time-based signal based derived from the ring RF, the time delay between the pump laser and the x-ray pulse was set to a variety of different delay times. To reduce the background contributions from the Compton scattering signal, a glancing exit detector geometry was used; the detector area to be integrated was varied during analysis to minimize the Compton contribution.

Result and discussion: Figure 2 shows typical experimental profiles for the XANES region. While the signal to noise was not optimal, the individual spectra were reproducible. То eliminate the need to recrystallize the sample, a specially sample designed sample structure was employed that takes advantage of the relatively large different in time scales of the amorphization and crystallization processes. By tuning the thickness of the more insulating (ZnS $-SiO_2$) layer, the heat trapped in the sample from the laser pulse re-crystallizes the initially reamorphized sample. As the time scale of the amorphization process is on the order of a nanosecond and the crystallization process is on the order of tens of nanoseconds, this unique thermal design allows access to both the amorphous and crystalline states during the same heating/cooling cycle. Figure 2 shows experimental XANES spectra for the Te K-edge (31.8 keV) taken for a variety of delays after the firing of the amorphization laser pulse. The bottom two traces show static measurement data for the liquid and laser re-amorphized phases, respectfully. The time evolution of the spectra from the amorphous state (1 ns) to that of the crystalline state (100 ms) can be visualized. It is notable that there is significantly more structure in the XANES features immediately after the amorphization pulse than can be seen in the static measurements of the liquid phase. This suggests that the highest temperature state may not be molten, but in fact may represent an excited state implying that at least for shorter laser pulses such at the 500 ps pulses used in the current experiment, there may be a non-thermal, electronic contribution the transformation

We are currently in the process of carrying out first-principle XANES simulations to compare to the experimental data. As the photoelectron energy for the XANES region is comparable to that of the covalent bonds present in the system,



Fig. 2. Experimental near-edge (XANES) spectra taken at the Te edge of a 50 nm GST film for various delay times after the amorphizing laser. Liquid XANES data obtained from separate experiments for the liquid phase of GST are shown for comparision. For reference, the bottom two spectra show data from static liquid and laser re-amorphized films, respectively.

the mean free scattering lengths are large (< 10nm) allowing probing of medium range order. The multiple scattering processes that are dominant in this region also contain significant information on the three dimensional nature of the local structure. We plan to publish this data in a peer reviewed journal when analysis is complete. We have already presented these results oral at the international conference XAFS-14 held in Camerino, Italy in July 2009.

References

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