

Epitaxial GeSbTe grown on Si(111)

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ABSTRACT

Epitaxial films of GeTe-Sb₂Te₃ alloys (GST) are deposited by molecular beam epitaxy (MBE) on a Si(111) substrate. The films studied by means of X-ray diffraction (XRD) reveal that the metastable cubic phase displays a single out-of plane orientation and the presence of an additional ordered structure incommensurate to the GST lattice periodicity. This structural ordering might be correlated with electronic ordering in the samples. We also performed fs-laser pulse excitation of GST that show that the low ps to fs time regime is the region of interest for the structural response of GST to fs-laser pulse excitation.

Key words: epitaxial GST, MBE, fs-laser pulse excitation

Epitaxial growth of GST was carried out on different substrates to obtain films with a single out-of-plane crystalline orientation. Depending on the chosen substrate temperature, four growth regimes exist, amorphous, polycrystalline, epitaxial, and no growth at all (sticking coefficients of the impinging species are zero). Metastable cubic GST grows with atomically flat surfaces only on (111)-oriented substrates.^{1,2}

At the actual stage of our growth tailoring and understanding we can predict that it is possible to achieve ordering in a GST alloy. In the previous studies we presented evidence that we can obtain layers with different alloy compositions located almost on the GeTe-Sb₂Te₃ pseudo-binary tie line, as if a “self-adjusting” mechanism existed. In fact, only by dramatically changing the impinging fluxes we were able to deviate from a composition lying along the pseudo-binary tie line. To shed light on this finding, we decided to investigate the two end points of the pseudo-binary tie line, GeTe and Sb₂Te₃. Interestingly, all epitaxial GeTe specimens present a composition of about Ge_{0.46}Te_{0.54}, which is off with respect to the perfectly stoichiometric GeTe films achieved by sputtering.³ An explanation could be the presence of vacancies in the Ge sub-lattice.

The results for GeTe imply that vacancies could also be present in epitaxial GST samples. In XRD measurements (specular ω -2 θ scans) we can clearly identify the peaks from the Si substrate and those related to GST. Interestingly, the scans exhibit additional peaks that stem neither from crystallographic orientations of GST other than (111) nor from phase separation. Strain effects were also ruled out. The origin of those peaks could be attributed to the presence of an incommensurate ordered structure in all the GST samples⁴. This structural ordering may in turn be correlated with electronic ordering in the samples. The analysis of the peaks reveals that the reiterative structure appears every six GST225 unit cells. However, careful theoretical simulations are under way to substantiate further the interpretation of these findings.

Importantly, the intensity ratio of the GST peak to the incommensurate peak varies as a function of composition: Both the peak shape and integral intensity are affected by changes in the supplied fluxes. This result may imply that the electronic ordering is influenced by the film composition, so

that it can be controlled by appropriately choosing the impinging fluxes and the growth temperature. This information is of paramount importance for tailoring the atomic order. This hypothesis is in accord with theoretical calculations and experimental results in which it was shown that the amount of vacancies and the degree of the structural Peierls distortions both change along the pseudo-binary tie line.⁵

It is important to note that our epitaxial layers could reversibly be switched back and forth between the SET and RESET states, highlighting the parallels and technological relevance to switching in polycrystalline films.⁶ In a recent study on the 100 ps timescale we have investigated the structural properties of as-grown epitaxial and fs laser-switched single-crystalline samples at the ESRF ID9B beamline using a laser pump (fs-laser irradiation) x-ray probe technique at an energy of 18keV (100 ps). We could show that x-ray diffraction clearly indicates that the low ps to fs time regime is the region of interest for the structural response of GST to fs-laser pulse excitation. A strong response of the diffraction pattern to the laser pump pulse -even far below the amorphization threshold is found on the ps time scale. The observed dynamics of the diffracted intensities are explained best by the excitation of coherent phonons in the GST film following laser pulse excitation. This leads to the proposition that switching from the crystalline to the amorphous phase occurs via the displacement of the atoms from their equilibrium positions. The displacement may occur via various pathways in a thermal or non-thermal transition, depending on the timescales and laser pulses applied to invoke the transition.

REFERENCES

¹ F. Katmis, R. Calarco, K. Perumal, P. Rodenbach, A. Giussani, M. Hanke, A. Proessdorf, A. Trampert, F. Grosse, R. Shayduk, R. Champion, W. Braun, and H. Riechert, *Crystal Growth & Design* **11**, 4606–4610 (2011).

² P. Rodenbach, R. Calarco, K. Perumal, F. Katmis, M. Hanke, A. Proessdorf, W. Braun, A. Giussani, A. Trampert, H. Riechert, P. Fons, and A. V. Kolobov, *Physica Status Solidi (RRL) - Rapid Research Letters* **6**, 415–417 (2012).

³ A. Giussani, K. Perumal, M. Hanke, P. Rodenbach, H. Riechert, and R. Calarco, *Physica Status Solidi (B)* **249**, 1939–1944 (2012).

⁴ W. Zhang, A. Thiess, P. Zalden, R. Zeller, P.H. Dederichs, J.-Y. Raty, M. Wuttig, S. Blügel, and R. Mazzarello, *Nature Materials* **11**, 952–6 (2012).

⁵ M. Wuttig and N. Yamada, *Nature Materials* **6**, 824–832 (2007).

⁶ P. Rodenbach, A. Giussani, K. Perumal, M. Hanke, M. Dubslaff, H. Riechert, R. Calarco, M. Burghammer, A.V. Kolobov, and P. Fons, *Applied Physics Letters* **101**, 061903 1–3 (2012).