Local Structure, X-ray Absorption and Localization Properties of GST and GeTe-based Phase Change Materials : an Ab Initio Simulation Approach.

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ABSTRACT

In the recent years, Ab Initio Density Functional Theory calculations have proven to be extremely useful in the production of structural models helping to interpret the output of many experiments, such as diffraction, x-ray absorption as well as other kind of spectroscopies.

In this work, we use a new implementation of XANES spectra calculations within DFT and PAW potentials to perform a fully ab initio calculation of these spectra, including the self-consistent treatment of the core-hole effects. We compute the site-resolved XANES signature of various GeTe and GST amorphous and crystal structures. These calculations show clearly the correlation between the near-edge features of the spectra and i) the local structural and chemical order inside the amorphous phase, and ii) with the distribution of the vacancies and cations in the metastable crystal phases. The calculations reproduce the main features of newly recorded XANES spectra for both the amorphous and crystalline phase allowing to obtain further information from the experiment, emphasizing the importance of the fluctuations and disorder. This approach provides a new interpretation of XANES measurements, relating essentially the phase change contrast to a moderate modification of the local environment of the Ge atoms.

On the other hand, the metastable crystal phase of GST materials has been shown recently to possess unique electron localization properties that can be tuned by an adequate annealing of the material [1]. Using DFT calculations, we show that the distribution of vacancies and cations inside Ge1Sb2Te4 can produce a variable localization of the states close to the Fermi level in agreement with new EXAFS measurements and the reported Anderson transition.

Finally, we study the effect of carbon incorporation inside the GeTe material. Strong carbon doping has been shown to increase significantly the recrystallization temperature of GeTe. Using DFT simulated annealing, we obtain a structural model that reproduces the trends measured by X-ray diffraction, showing the strong structuration of the amorphous phase through CGe4 units and long carbon chains.

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[1]. T. Siegrist et al., Nature Materials 10, 202 (2011)