

Development of phase change materials by density functional theory

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At present the discovery and development of new phase change materials is mainly based upon empirical strategies and trial- and error approaches [1,2,3]. Here we present an alternate approach, which is based upon ab initio calculations [4] of the ground state and the electronic band structure of binary and ternary Tellurium alloys. Density functional theory has been employed to determine the density of states, the band structure and the total energy for different structures of ternary alloys containing Ag, In, Sb and Te. The calculations show a variation of ground state structure and bonding type with stoichiometry. Understanding the observed trends promises the directed optimization of future phase change media. Comparing the theoretical predictions with experimental results of AgInTe_2 and AgSbTe_2 films it is demonstrated that only the AgSbTe_2 films which show a cubic coordination have sufficient density contrast and hence also optical contrast to allow phase change recording.

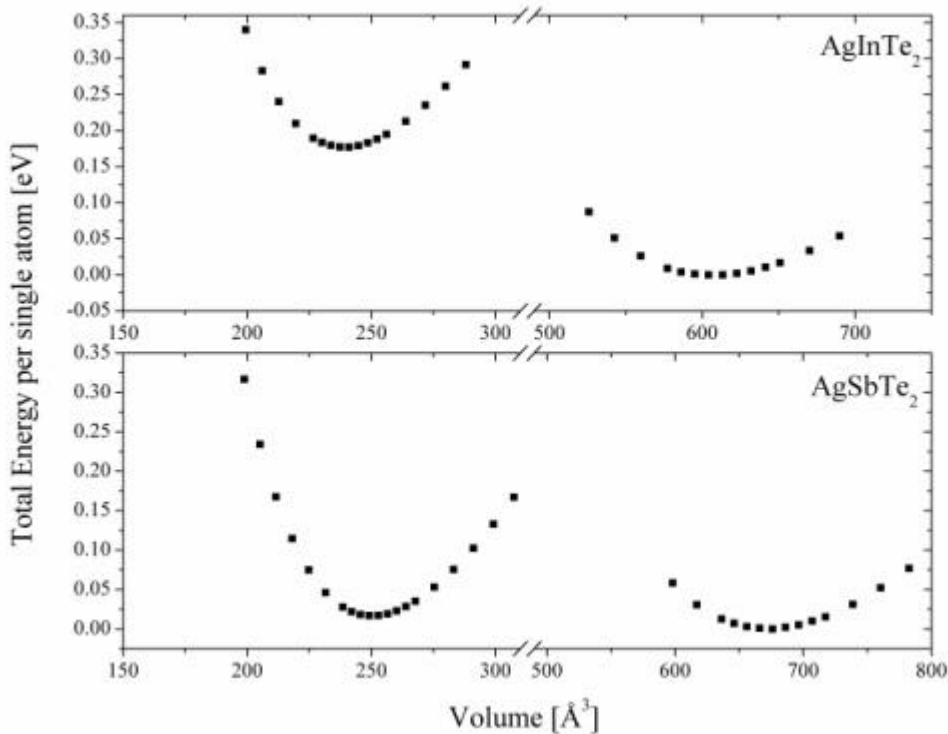


Figure 1: Variation of total energy per atom for different structures of AgInTe_2 and AgSbTe_2 .

Combinations of Ag, In, Sb and Te show as a function of their exact composition different crystallographic structures. We have used density functional theory in order to forecast the most stable configuration for different alloys, here AgInTe_2 and AgSbTe_2 , which are formed during crystallization processes of the commercially used AgInSbTe alloy. The variation of the total energy as a function of the unit cell volume for two common structures is shown in figure 1, where a rocksalt arrangement is depicted on the left hand side while the values for the unit cell on the right hand side correspond to the chalcopyrite structure. Adding entropical effects to these values, AgInTe_2 crystallizes in the chalcopyrite structure and the rocksalt configuration is preferred for AgSbTe_2 .

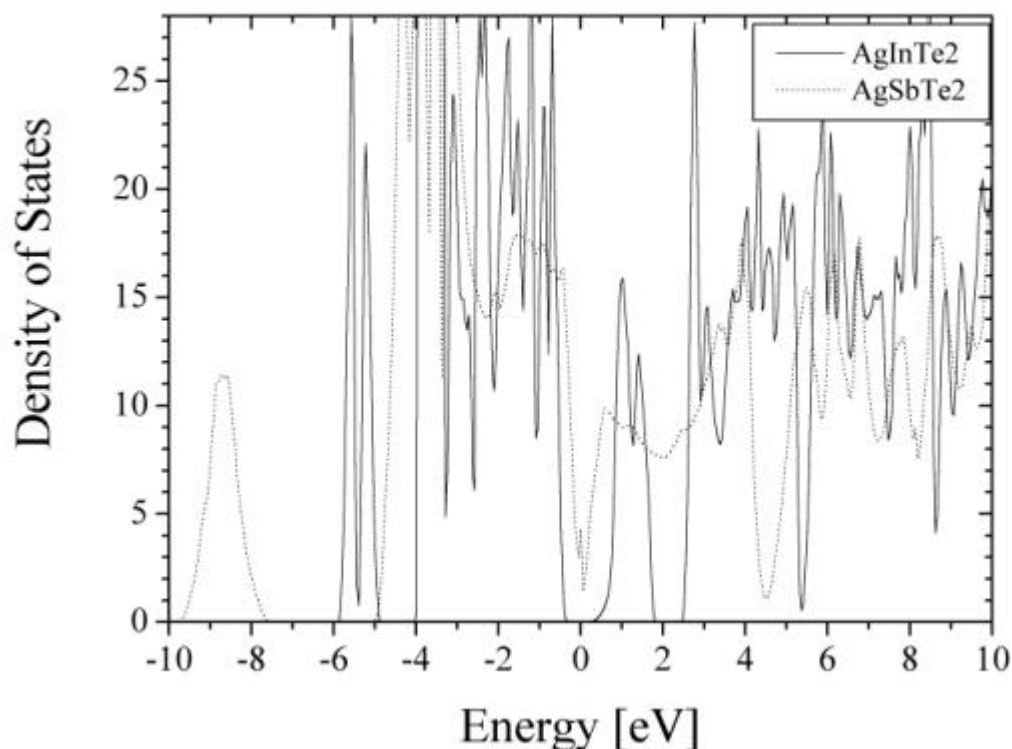


Figure 2: Theoretical density of states for the experimentally found structures of Chalcopyrite (AgInTe_2) and Rocksalt (AgSbTe_2).

The local arrangement of the atoms in the unit cell and in particular the different number of nearest neighbors affect the density of states, which is drawn in figure 2. The most important difference is the appearance of much broader structures for AgSbTe_2 due to the higher number of nearest neighbors ($Z=6$). For AgInTe_2 , where the coordination number Z is 4, much more localized states are observed. The structural change from the amorphous to the crystalline phase is accompanied by a change in density and thickness of the film, which is also closely linked to the crystalline structure. The different amount of density and thickness change can be clearly seen from figure 3, where a greater varying density is linked with a more pronounced shifted XRR spectra. Figure 4 shows the relationship between the increase in density respective decrease in thickness for both alloys and different annealing temperatures. The crystallization into the less close packed chalcopyrite structure leads to a smaller decrease in density, which is only 29% compared to the rocksalt structure of the AgSbTe_2 phase.

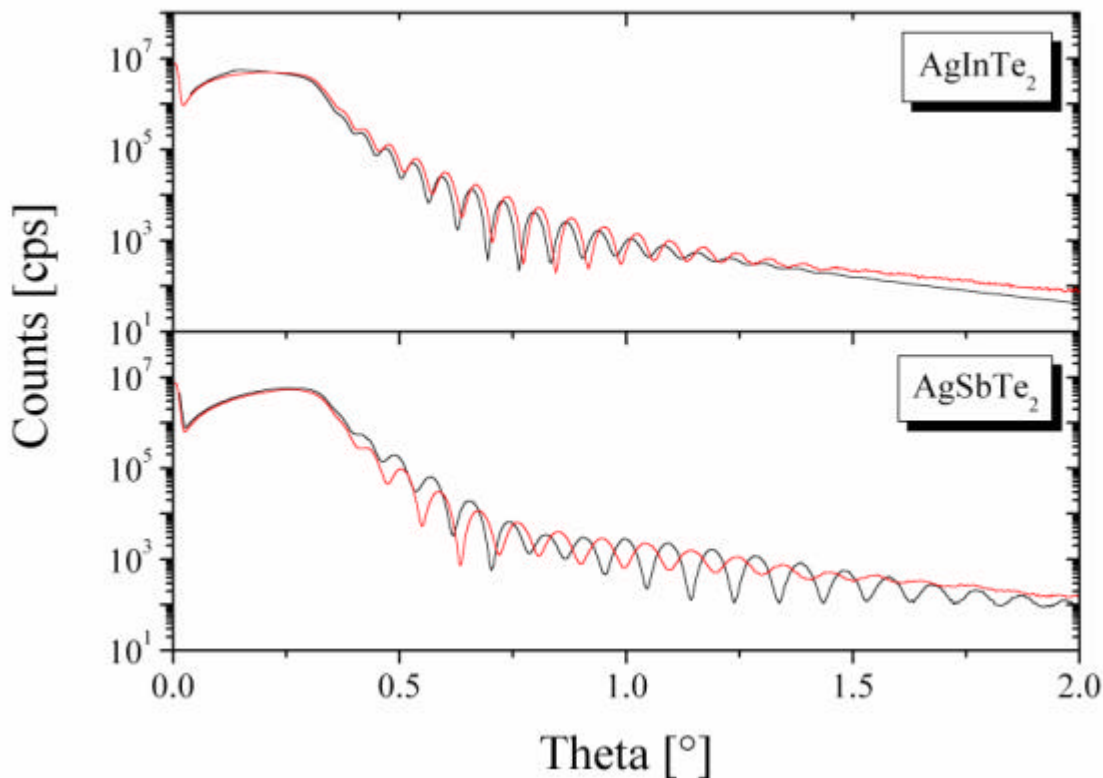


Figure 3: X-Ray reflectometry spectra of amorphous (black) and crystallized (red) AgInTe_2 and AgSbTe_2 .

This different extent of local rearrangement is also reflected in the evolution of the corresponding optical properties. Calculations of the normalized change of reflectance for a 60 nm thick film on a glass substrate show (figure 5), that the magnitude of the contrast change scales with the density change and so with the most stable crystal structure.

In summary we can conclude, that density functional theory is able to predict the stable structure for different alloys. Due to correlations between the local structure, the change of density and reflectivity upon crystallization, density functional theory can be employed as a useful tool for predicting feasible phase change materials.

Literature

- [1] M. Libera and M. Chen, MRS Bull. 15, 40 (1990)
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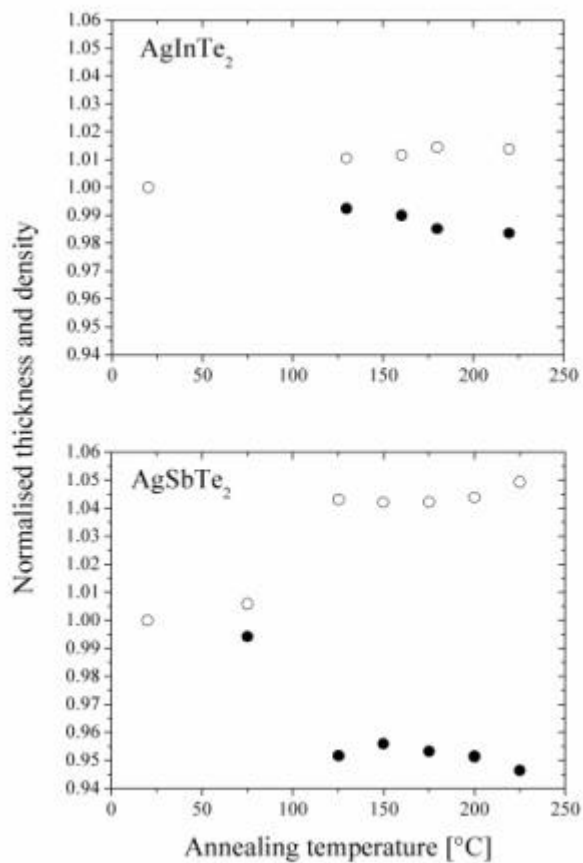


Figure 4: Evolution of density and thickness as function of temperature for both alloys. The structural change is more pronounced for AgSbTe_2 in contrast to AgInTe_2 .

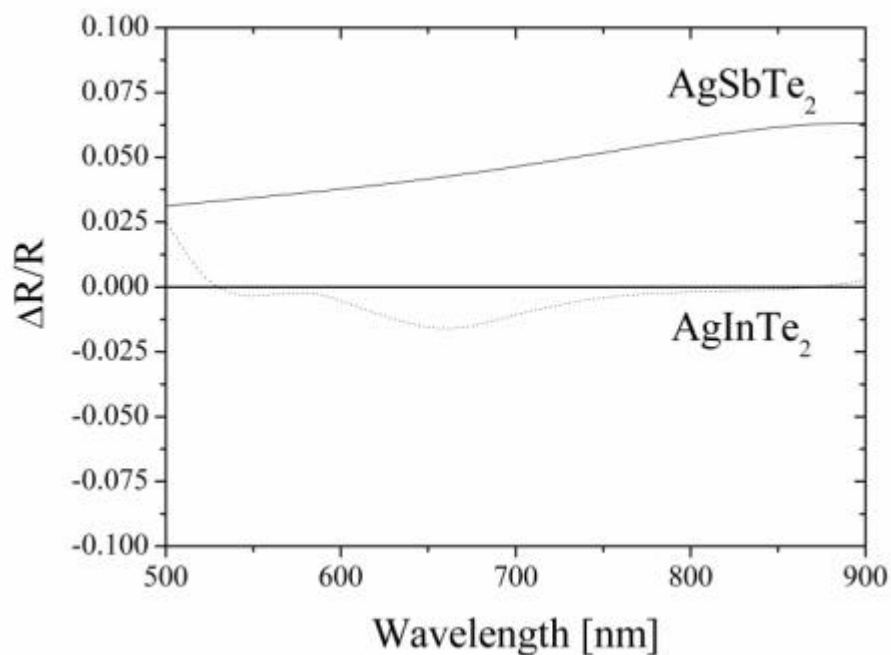


Figure 5: Normalised contrast for AgSbTe_2 and AgInTe_2 as a function of the laser wavelength. A thickness of 60nm on a glass slide was used in the model.