Ab initio-based simulations of electronic excitations in chalcogenide glasses.

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Abstract

Ab initio-based density-functional tight-binding (DFTB) molecular-dynamics computer silumations have been carried out to generate structural models of certain chalcogenide glasses (e.g. As_2S_3), and electronic excitations (e.g. as a result of optical illumination) in such models have been studied using spin-polarised geometry optimisation. The models reveal a number of interesting features, e.g. embedded, partially broken (As_4S_4) molecules, and a new type of coordination defect pair, namely a negatively-charged "see-saw" (trigonal prismatic) As_4^- defect and a near-planar S_3^+ defect which is stable in the electronic ground state and which is also created after electronic excitation of other defect configurations. In addition, we are investigating bond-breaking events following electronic (optical) excitation.