Structure determination with atomic-scale modeling of a new partially-disordered phase $AsTe_3$

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A new crystalline material with composition $AsTe_3$, recently obtained from crystallization of the parent $AsTe_3$ glass showed promising properties for thermoelectric applications.[1,2] The structure of this material could not be established directly from X-ray and neutron diffraction experiments, which exhibit broad peaks pointing to significant degrees of structural and/or chemical disorder. A semi-automatized protocol aiming to solve this structure with molecular modeling at the density functional level of theory (DFT), from the stand point of As/Te disorder, has been developed and implemented. The protocol is based on the Automated Interactive Infrastructure and Database for Computational Science (AiiDA),[3,4] a platform aiming to facilitate automatization, data storage and reproducibility, which offers in particular opportunities for the development of artificial intelligence in materials science modeling applications, such as the training of DFT-quality machine learning atomic interaction potentials.

Candidate $AsTe_3$ supercell structures derived from Te-metal and identified based on experimental diffraction and total scattering (a technique particularly adapted to the characterization of materials with structural disorder) are generated, evaluated, and progressively selected in a multi-step workflow comprising:

(i) combinatorial analyses of the repartitions of As and Te atoms with the supercell program[5] and corresponding aiida-supercell plugin,[6]

(ii) pre-evaluation of the structure stability from the stand point of electrostatic interactions (assuming the co-existence of metallic and more ionic Te species),

(iii) structure relaxations at the DFT level and progressively-increased accuracy settings, with and without constraints on the unit-cell from experimental DFT data.

The most stable $AsTe_3$ candidate structures obtained with this protocol are evaluated with respect to experimental data and compared to candidate structures obtained separately 'ex nihilo' with crystal structure prediction approaches employing genetic algorithms.[7,8] Further steps (in progress) will involve DFT-level molecular dynamics on best-candidate structures to account for the effects of local thermal vibrations on simulated total X-ray scattering and explore local structural configurations potentially representative of the structural disorder in this system. The semi-automatized protocols developed for this system will be largely transferrable to other systems, and contribute to enriching databases of calculated materials structures and properties.

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