DFT study of Si-Ge alloys: pressure-induced phases and vibrational signatures

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Abstract

Silicon–germanium (Si_xGe_{1-x}) alloys, long established in microelectronics, are experiencing renewed interest due to their versatile applications in both energy and information technologies. In particular, understanding their vibrational properties provides valuable insight into thermal transport, structural stability, and phonon-mediated functionalities in optoelectronic devices [1].

In this work, we investigate the structural and vibrational properties of Si_xGe_{1-x} alloys under pressure with x = 0.8, 0.5, 0.3 by combining in situ high-pressure Raman spectroscopy, X-ray diffraction experiments, and ab initio calculations within the framework of density functional theory (DFT), using the projector augmented-wave (PAW) scheme and density functional perturbation theory (DFPT) as implemented in VASP code [2]. The alloy was simulated using the genstr tool within the ATAT cluster expansion scheme with cluster sizes up to 16 atoms to ensure convergence [3]. To calculate the phonon frequencies, we used the post processing Phonopy package to analyze the vibrational modes [4]. High-pressure experiments reveal a pressure-induced transition from the cubic semiconducting phase to the metallic β-tin phase upon compression, while decompression leads to the coexistence of metastable r8 and bc8 phases at ambient conditions (Fig.1). This sequence is in excellent agreement with the results of the DFT calculations [5,6].

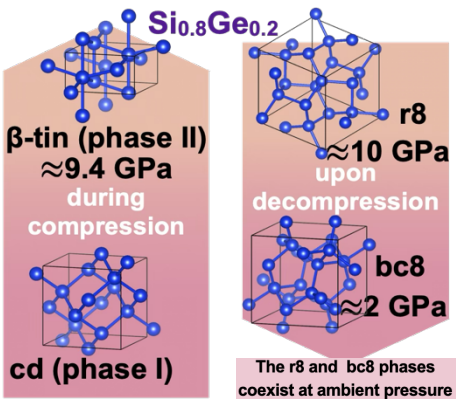

Fig.1: Pressure-induced phase transitions.

A key challenge in studying Si_xGe_{1-x} alloys is the interpretation of their Raman spectra, due to the lack of crystallographic symmetry in the disordered supercells used for phonon calculations. This makes it difficult to assign the calculated vibrational modes and to compare them with experimental results. In this study, we address this issue by applying a band unfolding approach based on projection operators, which consists of decomposing the vibrational modes of the disordered structures into the irreducible representations of an underlying ideal-ordered structure [7]. The peak positions in the resulting spectral function, which reflect the crystallographic symmetry, correspond to the phonon frequencies of the disordered system [5,8].

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