

Phonons of Mixed Crystals $\text{Cd}_{1-x}\text{Be}_x\text{Te}$ & $\text{Zn}_{1-x}\text{Mg}_x\text{S}$: Inelastic Neutron/Light (Raman) Scattering & *Ab Initio* Calculations Testing the Percolation Model

Abdelmajid Elmahjoubi

Abstract

The semiconductor mixed crystals $\text{A}_{1-x}\text{B}_x\text{C}$ with cubic (zinc-blende) or hexagonal (wurtzite) structures are of great interest for optoelectronics, as they offer the possibility to finely tune the bandgap and lattice parameters as a function of the composition x . More fundamentally, these alloys are the simplest disordered systems one can imagine. As such, they are ideal systems for studying the effects of disorder on physical properties, particularly the lattice dynamics. In this context, the percolation model was developed to explain how disorder influences the lattice dynamics. This model suggests that the vibration of a given chemical bond, $\text{A}-\text{C}$ or $\text{B}-\text{C}$, is sensitive to its local environment and can result in two distinct optical modes, depending on whether it vibrates in a 'same' or 'alien' environment. This behavior is intrinsic to random alloying. The percolation model has been successful in explaining the lattice dynamics of all cubic semiconductor alloys studied so far.

In this work, the percolation model is critically evaluated with respect to the vibrational behavior of two reference systems: zinc-blende $\text{Cd}_{1-x}\text{Be}_x\text{Te}$ and wurtzite $\text{Zn}_{1-x}\text{Mg}_x\text{S}$. Each of these systems presents a significant challenge to the model. The mixed crystal $\text{Cd}_{1-x}\text{Be}_x\text{Te}$ presents a stark contrast between the physical properties of its constituents, CdTe and BeTe . Here, we examine whether the percolation model still applies in such a high-contrast situation. The microscopic study of the mechanical/vibration properties of $\text{Cd}_{1-x}\text{Be}_x\text{Te}$ is completed by a mechanical study at the macroscopic scale concerned with the bulk modulus. Finally, the wurtzite $\text{Zn}_{1-x}\text{Mg}_x\text{S}$ mixed crystal is used to test how the percolation model, originally established to explain the vibrational behavior of cubic systems, transfers to low crystal symmetries. This test is crucial for validating the model's transferability and universality.

To explore the lattice dynamics of the studied mixed crystals experimental techniques and theoretical approaches were deployed. Raman scattering was performed under ambient conditions as well as under extreme temperature and pressure conditions. INS was carried out on large single crystals at the IN8 spectrometer (ILL). High-pressure XRD was done at the PSICHÉ beamline of the SOLEIL synchrotron (Paris). Finally, *ab initio* calculations were conducted using the DFT SIESTA code on large supercells generated with random substitution according to the special quasi-random structures (SQS) approach.

The two studied systems, despite their specific characteristics (strong contrast between the bond physical properties, and hexagonal symmetry), all exhibit a percolation-type behavior. This reinforces the status of the percolation model as a generic descriptor of the lattice dynamics of disordered mixed crystals.