Abstract

The semiconductor mixed crystals $A_{1-x}B_xC$ 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, A−C or B−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 three reference systems : zinc-blende $GaAs_{1-x}P_x$ and $Cd_{1-x}Be_xTe$, as well as wurtzite $Zn_{1-x}Mg_xS$. Each of these systems presents a significant challenge to the model. The first version of the model, which limits the sensitivity of vibrations to first-neighbor, has shown its limitations with $GaAs_{1-x}P_x$. The spectra of the imaginary part of the relative dielectric function $\Im\{\epsilon_r(\omega, x)\}$ (informing on the transverse optical modes) recently published by Zollner *et al.* [Appl. Phys. Lett. 123, 172102 (2023)] indicate that this approach does not faithfully describe the vibrational peak intensities. It is necessary to extend the sensitivity to second-neighbor in this system. The mixed crystal $Cd_{1-x}Be_xTe$ 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 $Cd_{1-x}Be_xTe$ is completed by a mechanical study at the macroscopic scale concerned with the bulk modulus. Finally, the wurtzite $Zn_{1-x}Mg_xS$ 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, covering phonons and phonon-polaritons, advanced experimental techniques and theoretical approaches were deployed. Raman scattering was performed under ambient conditions as well as under extreme temperature and pressure conditions. Inelastic neutron scattering was carried out on large single crystals at the IN8 beamline of the Institut Laue-Langevin (ILL) in Grenoble. High-pressure X-ray diffraction was done at the PSICHÉ beamline of the SOLEIL synchrotron in 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 three studied systems, despite their specific characteristics (sensitivity of vibrations to the local environment, 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.