Unidentified SiGe Phonon Mode: The effect of an atomic mass difference

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Introduction

The advancement of thermoelectric research involves accurate prediction of phonon behaviour and unravelling the mechanisms behind thermal transport. Accessing lower phonon frequency regions, however, is one of the more challenging limitations within this theme. Yokogawa *et al.* [1] successfully produced phonon spectra of bulk silicon-germanium (SiGe) alloy samples, from which the production of an unidentified phonon mode was observed. To gain further insight, this current study simulated SiGe alloys to test the effect of an atomic mass difference on the production of the unknown mode.

Simulation Methods

Cuboidal SiGe models with a cross-sectional diameter of 2.17 nm, and a length of 16.3 nm, were created to form SiGe alloys. The atomic configuration consisted of Si-Ge, Si-Si and Ge-Ge bonds, positioned randomly throughout the lattice. To test the effect of a mass difference, either the Si atomic mass was reduced, or the Ge atomic mass was increased, whilst keeping the potential uniform. The phonon dispersion relations were obtained by calculating the spectral energy density (SED), Φ, for a phonon mode, $(\boldsymbol{\kappa}, \omega)$, given by,

$$\Phi(\boldsymbol{\kappa},\omega) = \frac{1}{4\pi\tau_0 N_T}\sum_a\sum_b^B m_b \left| \int_0^{\tau_0} \sum_{n_{x,y,z}}^{N_T} \dot{u}_a\begin{pmatrix} n_{x,y,z} \\ b \end{pmatrix}; t\Big) \times exp\left[i\boldsymbol{\kappa}\cdot\boldsymbol{r}\begin{pmatrix} n_{x,y,z} \\ 0 \end{pmatrix} - i\omega t\right] dt \right|^2, \quad (1)$$

where τ_0 is the phonon lifetime, N_T total unit cells in the system, m_b the atomic mass of atom b, displaced, $\dot{u}_a$, in the direction a at a time t, $\boldsymbol{r}$ represents the equilibrium positions for each unit cell, $n_{x,y,z}$.

Results and Discussion

Reducing the Si mass caused the unknown mode, as well as the Si-Si optical band, to shift to higher frequencies. Contrastingly, increasing the Ge mass caused the unknown mode, and the Ge-Ge optical band, to shift to lower frequencies (Fig 1). Additionally, an increase in Ge mass caused a reduction in the unknown mode intensity (Fig 2a-b), but also reduced the frequency gap between the Ge-Ge optical mode and the unknown mode. The Si-Si and Ge-Ge band shifts but only when either the Si or Ge mass, respectively, changes (Fig 1), suggesting that there is not one element that influences the unknown mode, rather, both. The unknown mode only appears in the second Brillouin zone in the direction of kx, where the optical modes exist (Fig 2c). Although it is influenced by both Si and Ge, this mode exists only in the optical region, suggesting it is an optical like vibration mode.

Conclusion

Multiple SiGe alloys were simulated with varying Si or Ge atomic masses to test the effect on the intensity and frequency of the unknown mode. Reducing the Si or increasing the Ge atomic mass shifts the unknown mode to higher or lower frequencies, respectively, as well as the intensity. Further work entails testing the dependency of physical and mechanical parameters on the unknown mode.

Acknowledgements

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References

[1] R. Yokogawa, H. Takeuchi, Y. Arai, I. Yonenaga, M. Tomita, H. Uchiyama, T. Watanabe, A. Ogura, *Appl. Phys. Lett.*, **116**, 242104, (2020)

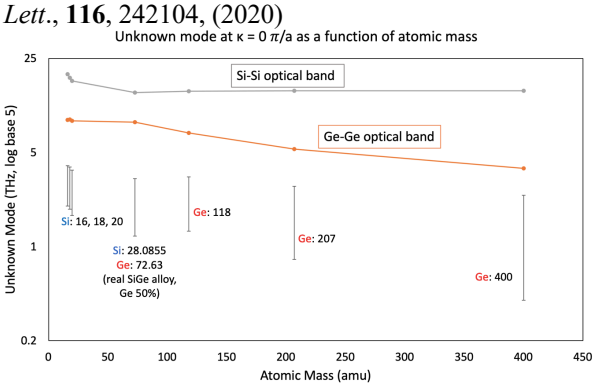

Figure 1: Plot showing the unknown mode frequency as a function of changing atomic mass. The points are labelled in accordance to what mass the respective element was changed to.

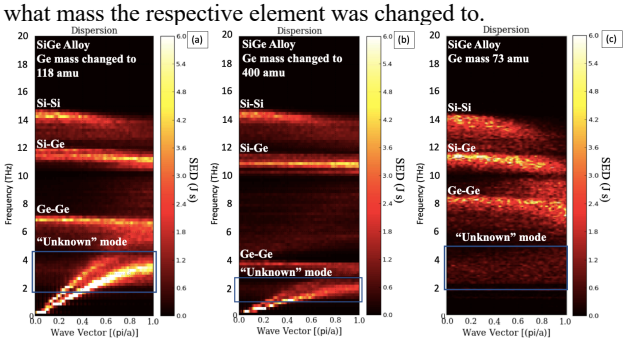

Figure 2 (a-c): Phonon dispersion relations depicting the SiGe alloy with a Ge mass of 118 amu (a), the SiGe alloy with a Ge mass of 400 amu (b) and (c) the extracted longitudinal optical (LO) mode dispersion relations with a Ge mass of 73 amu. The unknown mode is sectioned off by the blue rectangle.