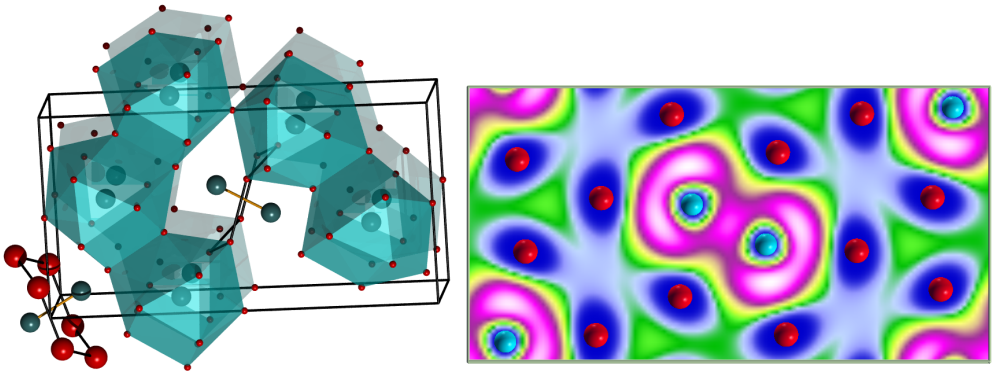
Rational Tuning and Thermodynamic Characterization of Lithium Silcides as Electrode Materials for Lithium Ion Batteries

Li_xSi may be a promising anode material for lithium ion batteries. We will discuss the crystal structures of the known stable phases and will identify similar structural elements (**fig. 1a**). In particular using the electron localization function (ELF) we will investigate the bonding in these structures (**fig. 1b**). The main goal of our investigation is to support an understanding of the charging and discharging processes, which are directly related to the Li^+ transport.



(a) Structure elements

(b) ELF plot: white high, blue low localization of electrons

Figure 1: Structure of $Li_{13}Si_4$ (red: lithium; blue: silicon)

For any application of Li_xSi as anode material a proper understanding of the thermodynamical behavior of the material is required. The calculation of the complete phonon dispersion as function of pressures allows for the construction of the Gibbs free energy, which then gives access to specific heat (**fig. 2**) or other thermodynamical data. The Gibbs free energy and derived thermodynamical quantities can be used for thermodynamical modeling of Li-ion batteries or as input for phase field simulations.

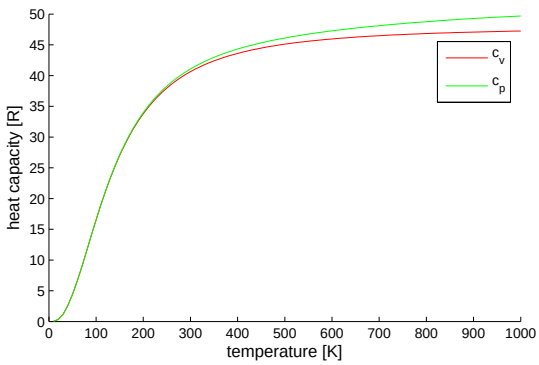


Figure 2: heat capacity of LiSi