Calculation of point defect formation free energy in multicomponent crystals

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Free energy of defect formation G_f is an important quantity that defines an equilibrium concentration of defects of a particular type. Usually G_f is calculated in molecular dynamics (MD) only at zero temperature, which is a trivial task. However, if a material is simulated close to its melting temperature, G_f can differ from the low-temperature value significantly, so the dependence $G_f(\mathrm{T})$ should be took into account. Calculations of this dependence are performed very rarely in MD, especially for multicomponent crystals [1, 2]. We have carried out the calculations for four materials: iron, aluminum, cooper and wüstite (FeO) in several different ways. The most simple and the least accurate of them is the calculation via the phonon spectra in the quasi-harmonic approximation [3]. In addition to it, we used three kinds of thermodynamic integration over a switching parameter λ [1, 4], one of them is original. Finally, thermodynamic integration along temperature was tested [2]. Agreement with papers [2, 4] was received, all methods were analyzed in detail, the most reliable and computationally effective one was selected.

[1] Fominykh N A, Nikolskiy V P and Stegailov V V 2023 *Comput. Mater. Sci.* **220** 112061
[2] Cheng B and Ceriotti M 2018 *Phys. Rev. B* **97** 054102
[3] Fultz B 2010 *Prog. Mater. Sci.* **55** 247–352
[4] Korotaev P, Belov M and Yanilkin A 2018 *Comput. Mater. Sci.* **150** 47–53