

Thermodynamics of Materials through first-principles molecular dynamics and the Reversible Scaling method

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One of the most challenging problems in computational condensed matter physics is the calculation of the thermodynamic properties of materials by simulation methods. The calculation of free energy and its temperature dependence based on DFT [as recently performed by *Alfè et al., IJQC 77 871 (2000)*], implies a high computational cost, since a complete equilibrium simulation has to be performed at each temperature. More efficient techniques have been proposed to obtain free energies, especially the Reversible Scaling (RS) method [*Koning et al., PRL 83 3973 (1999)*], which allows an efficient and accurate determination of the free energy over a wide range of temperatures (or other thermodynamical parameters) using a single simulation and including all anharmonic effects. The thermodynamics of various phases of silicon (crystalline, amorphous and liquid, as well dislocations and clathrate structures) have been studied using RS within Monte Carlo simulations and empirical potentials [*Miranda and Antonelli, 26th ICPS - 2002*]. In this work, we show an implementation of the Reversible Scaling method within first-principles molecular dynamics using the SIESTA method for linear-scaling DFT. This new methodology allows an efficient way to determine the thermodynamics of materials as a function of temperature based on first principles calculations. We present the main ideas of this methodology and some recent tests and applications.